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Jc772 U.S. PTO

# UTILITY PATENT APPLICATION TRANSMITTAL

(Only for new nonprovisional applications under 37 CFR 1.53(b))

Attorney Docket No.	35.C14598
First Named Inventor or Application Identifier	
HIDEO UKUDA	
Express Mail Label No.	

Jc714 U.S. PTO  
09/606522  
06/30/00

## APPLICATION ELEMENTS

See MPEP chapter 600 concerning utility patent application contents.

**ADDRESS TO:** Assistant Commissioner for Patents  
Box Patent Application  
Washington, DC 20231

1. <input checked="" type="checkbox"/> Fee Transmittal Form (Submit an original, and a duplicate for fee processing)	6. <input type="checkbox"/> Microfiche Computer Program (Appendix)
2. <input checked="" type="checkbox"/> Specification Total Pages <b>65</b>	7. Nucleotide and/or Amino Acid Sequence Submission (if applicable, all necessary)
3. <input checked="" type="checkbox"/> Drawing(s) (35 USC 113) Total Sheets <b>13</b>	a. <input type="checkbox"/> Computer Readable Copy
4. <input checked="" type="checkbox"/> Oath or Declaration Total Pages <b>1</b>	b. <input type="checkbox"/> Paper Copy (identical to computer copy)
a. <input type="checkbox"/> Newly executed (original or copy)	c. <input type="checkbox"/> Statement verifying identity of above copies
b. <input checked="" type="checkbox"/> Unexecuted for information purposes	
c. <input type="checkbox"/> Copy from a prior application (37 CFR 1.63(d)) (for continuation/divisional with Box 17 completed) <b>[Note Box 5 below]</b>	
i. <input type="checkbox"/> <b>DELETION OF INVENTOR(S)</b> Signed Statement attached deleting inventor(s) named in the prior application, see 37 CFR 1.63(d)(2) and 1.33(b).	
5. <input type="checkbox"/> Incorporation By Reference (useable if Box 4c is checked) The entire disclosure of the prior application, from which a copy of the oath or declaration is supplied under Box 4c, is considered as being part of the disclosure of the accompanying application and is hereby incorporated by reference therein.	

## ACCOMPANYING APPLICATION PARTS

8. <input type="checkbox"/> Assignment Papers (cover sheet & document(s))	
9. <input type="checkbox"/> 37 CFR 3.73(b) Statement (when there is an assignee)	<input type="checkbox"/> Power of Attorney
10. <input type="checkbox"/> English Translation Document (if applicable)	
11. <input type="checkbox"/> Information Disclosure Statement (IDS)/PTO-1449	<input type="checkbox"/> Copies of IDS Citations
12. <input checked="" type="checkbox"/> Preliminary Amendment	
13. <input checked="" type="checkbox"/> Return Receipt Postcard (MPEP 503) (Should be specifically itemized)	
14. <input type="checkbox"/> Small Entity <input type="checkbox"/> Statement filed in prior application Statement(s) Status still proper and desired	
15. <input type="checkbox"/> Certified Copy of Priority Document(s) (if foreign priority is claimed)	
16. <input type="checkbox"/> Other:	

17. If a CONTINUING APPLICATION, check appropriate box and supply the requisite information:  
☐ Continuation    ☐ Divisional    ☐ Continuation-in-part (CIP)    of prior application No. \_\_\_\_\_

## 18. CORRESPONDENCE ADDRESS

<input checked="" type="checkbox"/> Customer Number or Bar Code Label	<b>05514</b> (Insert Customer No. or Attach bar code label here)	or <input type="checkbox"/> Correspondence address below
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CLAIMS	(1) FOR	(2) NUMBER FILED	(3) NUMBER EXTRA	(4) RATE	(5) CALCULATIONS
	TOTAL CLAIMS (37 CFR 1.16(c))	195-20 =	175	X \$ 18.00 =	\$ 3150.00
	INDEPENDENT CLAIMS (37 cfr 1.16(b))	5-3 =	2	X \$ 78.00 =	\$ 156.00
	MULTIPLE DEPENDENT CLAIMS (if applicable) (37 CFR 1.16(d))			\$ 260.00 =	\$ 260.00
				BASIC FEE (37 CFR 1.16(a))	\$ 690.00
			Total of above Calculations =		\$ 4256.00
	Reduction by 50% for filing by small entity (Note 37 CFR 1.9, 1.27, 1.28).				
	TOTAL =				\$ 4256.00

19. Small entity status

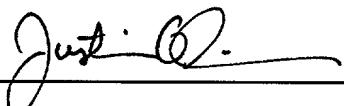
- a. ☐ A Small entity statement is enclosed
- b. ☐ A small entity statement was filed in the prior nonprovisional application and such status is still proper and desired.
- c. ☐ Is no longer claimed.

20. ☒ A check in the amount of \$ 4256.00 to cover the filing fee is enclosed.

21. ☐ A check in the amount of \$ \_\_\_\_\_ to cover the recordal fee is enclosed.

22. The Commissioner is hereby authorized to credit overpayments or charge the following fees to Deposit Account No. 06-1205:

- a. ☒ Fees required under 37 CFR 1.16.
- b. ☐ Fees required under 37 CFR 1.17.
- c. ☐ Fees required under 37 CFR 1.18.

SIGNATURE OF APPLICANT, ATTORNEY, OR AGENT REQUIRED	
NAME	Justin J. Oliver - Reg. No. 44,986
SIGNATURE	
DATE	June 30, 2000

JJO\cmv

35.C14598

PATENT APPLICATION

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of: )  
HIDEO UKUDA ) Examiner: Unassigned  
Application No.: Unassigned ) Group Art Unit: Unassigned  
Filed: Concurrently )  
For: OPTICAL MATERIAL AND )  
OPTICAL SYSTEM USING IT : June 30, 2000

Commissioner for Patents  
Washington, D.C. 20231

PRELIMINARY AMENDMENT

Sir:

Prior to examination on the merits, please amend  
the above-identified application as follows:

IN THE CLAIMS:

Please amend Claims 27, 29 and 31, as follows:

27. (Amended) An optical member comprising the  
optical material as set forth in [either] any one of Claims  
[1 to 26] 1-11 and 13-26.

29. (Amended) A diffracting optical element using  
the optical material as set forth in [either] any one of  
Claims [1 to 26] 1-11 and 13-26.

31. (Amended) An optical device comprising the optical system of Claim 28 [or 30].

Please add new Claims 37-43, as follows:

--37. An optical device comprising the optical system of Claim 30.

38. An optical member comprising the optical material as set forth in Claim 12.

39. An optical system comprising the optical member of Claim 38.

40. A diffracting optical element using the optical material as set forth in Claim 12.

41. An optical system comprising the diffracting optical element of Claim 40.

42. An optical device comprising the optical system of Claim 39.

43. An optical device comprising the optical system of Claim 41.--.

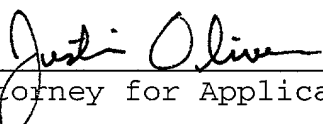
REMARKS

Claims 1-43 are now pending in this application, with Claims 1, 10, 11, 17 and 32 being independent. By this Amendment, Applicant has amended Claims 27, 29 and 31, and added new Claims 37-43. The changes merely correct formal matters regarding the multiple dependent claims.

Applicant requests favorable consideration and early passage to issue of the present application.

Applicant's undersigned attorney may be reached in our Washington, D.C. office by telephone at (202) 530-1010. All correspondence should continue to be directed to our below-listed address.

Respectfully submitted,

  
\_\_\_\_\_  
Attorney for Applicant  
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JJO/tmm



For the purpose of correction for this chromatic aberration, there are suggestions on methods of controlling the refractive index and Abbe's number in order to expand the range of the optical constants of glass materials, thereby obtaining the low-refraction high-dispersion glass materials, for example, in Japanese Patent Application Laid-Open No. 6-32631, Japanese Patent Application Laid-Open No. 61-9262, Japanese Patent Publication No. 4-33740, and so on.

On the other hand, other references, e.g. SPIE Vol. 1354 International Lens Design Conference (1990) etc., disclose methods of reducing the chromatic aberration by use of a diffracting optical element provided with a diffraction grating having diffracting action in part of a lens surface or an optical system, in contrast to the methods of reducing the chromatic aberration by the combination of glass materials.

These methods make use of the physical phenomenon in which a refracting surface and a diffracting surface in the optical system demonstrate opposite directions of occurrence of chromatic aberration against rays of a certain reference wavelength.

Further, such diffracting optical element can be provided with an aspherical-lens-like effect by changing periods of the periodic structure of the diffraction grating and is thus greatly effective to reduction of aberration.

Comparing them herein as to refraction of rays, a ray appears as one ray even after refracted by a lens surface, whereas a ray appears as a plurality of rays of plural orders after diffracted by a diffractive surface.

When the diffracting optical element is used as a lens system, it is thus necessary to determine the grating structure so as to concentrate light of the used wavelength range into specific orders (which will also be referred to hereinafter as "designed orders") by adequately enhancing the diffraction efficiency of rays of the designed orders. When light is concentrated in the specific orders, the intensity of rays of diffracted light of the other orders is low. When the intensity is zero, diffracted light thereof appears null. If there exist rays of diffraction orders other than the designed orders, they will be focused at positions different from those of the rays of the designed orders and appear as flare light.

The layouts capable of reducing this decrease of diffraction efficiency are presented in Japanese Patent Application Laid-Open No. 9-127321 and Japanese Patent Application Laid-Open No. 11-44808. The layouts realize high diffraction efficiency in a wide wavelength range by optimizing dispersion properties and thicknesses of gratings of materials different from each other.

Specifically, they disclose the diffracting optical elements wherein a plurality of optical materials (layers) are stacked on a substrate and wherein the diffraction grating is formed in the step shape, kinoform, or binary shape resulting from approximation thereof to the step shape, or the like in at least one of interfaces between the mutually different optical materials.

In each of the diffracting optical elements described in Japanese Patent Application Laid-Open No. 9-127321 and Japanese Patent Application Laid-Open No. 11-44808, the combination of a relatively high-refraction low-dispersion material with a relatively low-refraction high-dispersion material is used in order to obtain the layout having the high diffraction efficiency in the wide wavelength range. Specifically, the materials used are as follows: BMS81 ( $n_d = 1.64$ ,  $v_d = 60.1$ : available from Ohara Inc.) and the plastic optical material PC ( $n_d = 1.58$ ,  $v_d = 30.5$ : available from Teijin Chemicals Ltd.) in the case of Japanese Patent Application Laid-Open No. 9-127321; C001 ( $n_d = 1.5250$ ,  $v_d = 50.8$ : available from Dainippon Ink and Chemicals, Inc.), the plastic optical material PC ( $n_d = 1.58$ ,  $v_d = 30.5$ : available from Teijin Chemicals Ltd.), BMS81 ( $n_d = 1.64$ ,  $v_d = 60.1$ : available from Ohara Inc.) etc. in the case of Japanese Patent Application Laid-Open No. 11-44808.

For further enhancing the optical performance of the aforementioned diffracting optical elements, we investigated the optical materials commercially available, or the optical materials under research and development and obtained the distribution as illustrated in Fig. 1.

The materials of the stack diffracting optical elements described in the Japanese Patent Applications Laid-Open No. 9-127321 and Laid-Open No. 11-44808 also fall in the distribution of Fig. 1.

As for the materials suggested in the aforementioned Japanese Patent Applications Laid-Open No. 6-32631 and Laid-Open No. 61-9262 and Japanese Patent Publication No. 4-33740, their refractive indexes and refractive-index dispersions are ( $n_d = 1.585$  to  $1.660$ ,  $v_d = 40.5$  to  $32.5$ ), ( $n_d = 1.5945$  to  $1.6925$ ,  $v_d = 27.3$  to  $36.6$ ), and ( $n_d = 1.55$  to  $1.65$ ,  $v_d = 27$  to  $35$ ), respectively, and these materials also fall in the distribution of the existing substances of Fig. 1.

It is seen that 2-ethoxy-ethyl methacrylate ( $n_d = 1.483$ ,  $v_d = 32$ ) seems only one material with low refractive index and high dispersion and that there have been few such materials developed.

## SUMMARY OF THE INVENTION

An object of the present invention is to provide an optical material with low refractive index and high

dispersion, which was not known heretofore, and an optical system using it.

A first aspect of the present invention is an optical material which is a mixture of materials comprising a first material having a refractive index of not more than 1.45 for the d-line and a second material having an Abbe's number, indicating wavelength dispersion in the visible region, of not more than 25, wherein a relation between the refractive index for the d-line ( $n_d$ ) and the Abbe's number ( $v_d$ ) indicating the wavelength dispersion in the visible region is defined as follows:

$$n_d \leq -6.667 \times 10^{-3} v_d + 1.70.$$

A second aspect of the invention is the optical material of the first aspect wherein said Abbe's number ( $v_d$ ) indicating the wavelength dispersion in the visible region is less than 40.

A third aspect of the invention is the optical material of the first aspect or the second aspect wherein said second material comprises particles having the grain size in the range of 2 to 100 nm.

A fourth aspect of the invention is the optical material of the first aspect, the second aspect, or the third aspect wherein said first material is an amorphous fluororesin.

A fifth aspect of the invention is the optical material of either one of the first to fourth aspects







A nineteenth aspect of the invention is the optical material of the seventeenth aspect or the eighteenth aspect wherein said second material comprises particles having the grain size in the range of 2 to 100 nm.

A twentieth aspect of the invention is the optical material of the seventeenth aspect, the eighteenth aspect or the nineteenth aspect wherein said second material is ITO (indium-tin-oxide).

A twenty first aspect of the invention is the optical material of either one of the seventeenth to twentieth aspects wherein said first material is polymethyl methacrylate.

A twenty second aspect of the invention is the optical material of either one of the seventeenth to twenty first aspects wherein said first material is polymethyl methacrylate, said second material is particles of ITO (indium-tin-oxide), and a weight ratio of the particles and said polymethyl methacrylate is in the range of 30:100 to 250:100.

A twenty third aspect of the invention is the optical material of either one of the seventeenth to twentieth aspects wherein said first material is an amorphous polyolefin.

A twenty fourth aspect of the invention is the optical material of either one of the seventeenth to twentieth aspects wherein said first material is an







Fig. 11 is a diagram to show the relationship between refractive index and dispersion in Comparative Example 2;

Fig. 12 is a diagram to show the relationship between refractive index and dispersion in Example 4, Comparative Example 4, and Comparative Example 5;

Fig. 13 is a diagram to show the relationship between refractive index and dispersion in Example 5 and Example 6; and

Fig. 14 is a diagram to show the relationship between refractive index and dispersion in Example 8.

#### DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

Embodiments of the optical materials according to the present invention will be described below.

In general, since a substance is composed of molecules or atoms, oscillator characteristics and density are determined by its atomic or molecular structure. For this reason, the optical constants, including the refractive index  $n_d$  for the d-line (light of the wavelength 587 nm), the Abbe's number  $v_d$ , indicating wavelength dispersion in the visible region, etc. of the optical materials, show the distribution as illustrated in Fig. 1. The relation between refractive index  $n$  and polarization  $\chi'$  of substance can be expressed by the following Equation (1).

$$n^2 = 1 + \chi' \quad (\text{Eq. 1})$$

Supposing particles have the size of 2 to 100 nm, the polarization characteristics inside the particles will be like those of bulk. However, in the case of a structure with a reduced filling factor of substance as illustrated in Fig. 2, the polarization characteristics for the light in the visible wavelength range of 400 to 700 nm are of a level in which nonuniformity thereof can be ignored in an optical sense. Therefore, the structure as illustrated in Fig. 2 demonstrates the optical characteristics close to the Drude theory of Eq. (2) below.

$$n^2 = 1 + \chi^1 = 1 + T(n_1^2 - 1) \quad (\text{Eq. 2})$$

$(0 \leq T \leq 1)$   $n_1$ : refractive index of bulk

Fig. 3 is an explanatory diagram to show the relationship between refractive index  $n_d$  and Abbe's number  $v_d$  with various filling factors of substance. As seen from Fig. 3, the refractive index  $n_d$ , and dispersion  $v_d$  vary as indicated by curves S1, S2, S3, and with variation in apparent density by changing the filling factors, the refractive index demonstrates change while the Abbe's number little change. Therefore, a substance can be prepared in the region of  $(n_d \leq -6.667 \times 10^{-3}v_d + 1.70, v_d \leq 40, \text{ and } n_d \leq -0.01v_d + 1.70, v_d \leq 40)$  by decreasing the refractive index in the structure of Fig. 2.

When an antireflection film is formed on a surface of a lens by vapor deposition or the like, the filling factor of the film is lowered depending upon conditions of deposition and the structure becomes as illustrated in Fig. 2, whereby the refractive index of the material with large wavelength dispersion (small Abbe's number  $v_d$ ) is decreased to some extent.

However, if the filling factor of the substance is too low as in (2) of Fig. 4, there will appear such a tendency that when the substance is exposed from vacuum to the ordinary atmosphere, molecules of air and water adsorb thereto because of its porous structure, so as to impede the decrease of refractive index and increase the Abbe's number as well. In addition, the substance becomes very fragile.

Therefore, a material with a low refractive index and high dispersion (small Abbe's number) is realized by first decreasing the filling factor of the substance with large wavelength dispersion of refractive index (hereinafter referred to as "dispersion") (a high-dispersion substance) and filling gaps in this substance with another low-index substance with a small refractive index, as in (3) of Fig. 4.

Refractive indexes and Abbe's numbers were calculated according to Eq. (3) below and based on the polarizability obtained by Eq. (1), for mixtures based on the assumption that the material with high



the size is in units not more than 100 nm).

Specifically, the following mixing ways are considered, as illustrated in Fig. 6;

where the high-dispersion material is of fine  
5 particles,

(1) normal mixing;

where the main chain is an organic polymer,

(2) guest-host type mixing,

(3) main chain type mixing, and

10 (4) side chain type mixing.

In the case of the film formation by vacuum vapor deposition, applicable methods include two-dimensional evaporation of the large-dispersion material and the low-index material, formation of equivalent layers by  
15 depositing the respective materials in units of several nm, and so on.

However, as the units indicating high dispersion of polarizability become 2 nm or less to go into the molecular level, the environment of the high-dispersion  
20 units becomes far from the bulk state and tends to follow the Lorentz-Lorenz theory (Eq. (4)) with consideration to local electric fields, rather than the Drude theory. In this case, where the materials are mixed in fixed proportions, the dispersion tends to be  
25 lower, as compared with Eq. (3).

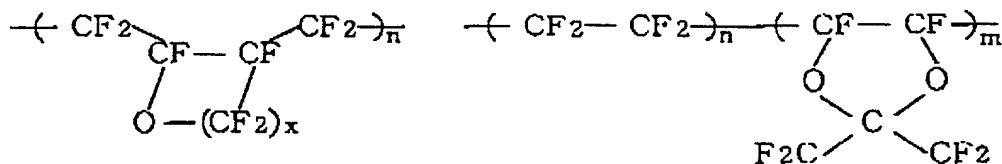
$$n^2 = [\{C_1 n_1^2 / (\rho_1 (n_1^2 + 2))\} + \{C_2 n_2^2 / (\rho_2 (n_2^2 + 2))\}] / [\{C_1 / (\rho_1 (n_1^2 + 2))\} + \{C_2 / (\rho_2 (n_2^2 + 2))\}] \quad (\text{Eq. 4})$$

$C_x$ : weight concentration,

$n_i$ : refractive index of bulk of each material.

$\rho_x$ : density in bulk of each material

The blending materials can be either organic or inorganic materials if they are the material having large dispersion and the material indicating the property of low refractive index. There are no specific restrictions on the materials, but specific examples of the materials are the following materials. For the material with the low refractive index ( $n_d \leq 1.45$ ), examples of the organic materials are amorphous fluororesins ( $n_d = 1.28$  to  $1.38$ ) represented by the following chemical formula, for example.



Examples of the inorganic materials are  $\text{MgF}_2$  ( $n_d = 1.38$ ),  $\text{CaF}_2$  ( $n_d = 1.44$ ), and so on.

There are no specific restrictions on the large-dispersion material ( $v_d \leq 25$ ), but specific examples thereof are  $\text{TiO}_2$  ( $n_d$ ,  $v_d = 2.2652$ , 11.8),  $\text{Nb}_2\text{O}_5$  ( $n_d$ ,  $v_d = 2.367$ , 14.0), ITO ( $n_d$ ,  $v_d = 1.8581$ , 5.53),  $\text{Cr}_2\text{O}_3$  ( $n_d$ ,  $v_d =$

2.2178, 13.4),  $\text{BaTiO}_3$  ( $n_d$ ,  $v_d = 2.4362, 11.3$ ), etc. and mixtures containing at least one of the forgoing materials.

5 The optical material in the region of ( $n_d \leq -6.667 \times 10^{-3}v_d + 1.70$ ,  $v_d \leq 40$ ) can be realized by using the low-index material of  $n_d \leq 1.45$  and particles of the large-dispersion material of  $v_d \leq 25$  in the grain size of 2 to 100 nm.

10 There are no specific restrictions on the low-index material, but it is desirably one of the amorphous fluororesins. The reason is that the amorphous fluororesins have low refractive indexes, are stable and less reactive, and give rise to less deviation from calculated values in the form of  
15 mixtures.

There are no specific restrictions on the material with large dispersion (i.e., with a small Abbe's number), but it is desirably particles of a composite metal oxide of titanium and silicon ( $\text{Si}_x\text{-Ti}_{(1-x)}\text{O}_2$ ). The  
20 ratio of metals can be adjusted to an arbitrary value for the composite metal oxide of titanium and silicon and it is thus easy to adjust the Abbe's number. The large-dispersion material used herein is the particles of the composite metal oxide of titanium and silicon  
25 adjusted to the Abbe's number ( $v_d$ ) of  $24.5 \leq 25$ .

There are no specific restrictions on the grain size of the particles of the composite metal oxide of

Ti and Si ( $\text{Si}_x\text{-Ti}_{(1-x)}\text{O}_2$ ) as long as it does not cause unignorable scattering of light. The grain size is desirably not more than 100 nm.

For uniformly dispersing the above particles of ( $\text{Si}_x\text{-Ti}_{(1-x)}\text{O}_2$ ), for example, in the above amorphous fluororesins, it is effective to modify the surface of the particles of ( $\text{Si}_x\text{-Ti}_{(1-x)}\text{O}_2$ ) by a fluorophilic group like the fluoroalkyl group.

When the mixing ratio of the fluororesin and the composite metal oxide of ( $\text{Si}_x\text{-Ti}_{(1-x)}\text{O}_2$ ) is in the range of 45:100 to 75:100, the resultant optical material is one satisfying the region ( $n_d \leq -6.667 \times 10^{-3}v_d + 1.70$ ,  $v_d \leq 40$ ).

As described above, the optical material in the region ( $n_d \leq -6.667 \times 10^{-3}v_d + 1.70$ ,  $v_d \leq 40$ ) is obtained by mixing the material with the low index ( $n_d \leq 1.45$ ) and the material with the large wavelength dispersion of refractive index ( $v_d \leq 25$ ).

In the present embodiment the low-index high-dispersion optical material is realized as described above.

The aforementioned material, 2-ethoxy-ethyl methacrylate, is considered to be a material realizing ( $n_d = 1.483$ ,  $v_d = 32$ ) by decreasing the density in terms of the structure, but the structure of this 2-ethoxy-ethyl methacrylate is different from that of the optical material in the region ( $n_d \leq -6.667 \times 10^{-3}v_d +$

1.70,  $v_d \leq 40$ ) prepared by forming the mixture of the material with the low index ( $n_d \leq 1.45$ ) and the large-dispersion material ( $v_d \leq 25$ ) in the present embodiment. In addition, the method with this 2-ethoxy-ethyl methacrylate provides less freedom for adjusting the levels of refractive index and wavelength dispersion than the method of the present embodiment.

As another example, the optical material is realized in the region of ( $n_d \leq -0.01v_d + 1.70$ ,  $v_d \leq 40$ ) by using the low-index material of  $n_d \leq 1.40$  and particles with  $v_d \leq 15$  and in the particle size of 2 to 100 nm as the large-dispersion material.

There are no specific restrictions on the low-index material for realizing this optical material, but the low-index material is desirably one of the amorphous fluororesins. The reason is that the amorphous fluororesins have the low refractive indexes, are stable and less reactive, and thus give rise to less deviation from calculated values in the form of mixtures.

There are no specific restrictions on the large-dispersion material for realizing the above optical material, but it is desirably particles of  $\text{TiO}_2$ . The reason is that the  $\text{TiO}_2$  particles are relatively easily available and belong to the class of materials indicating the highest dispersion out of the particles for optical use commercially available commonly.

There are no specific restrictions on the particle size of the  $\text{TiO}_2$  particles as long as it does not cause unignorable scattering of light. The particle size is preferably not more than 100 nm.

5 For uniformly dispersing the above  $\text{TiO}_2$  particles, for example, in the aforementioned amorphous fluororesins, it is effective to modify the surface of the  $\text{TiO}_2$  particles by a fluorophilic group like the fluoroalkyl group.

10 When the mixing ratio of the fluororesin and titanium oxide ( $\text{TiO}_2$ ) is in the range of 7:100 to 90:100, the optical material obtained satisfies the region ( $n_d \leq -6.667 \times 10^{-3}v_d + 1.70$ ,  $v_d \leq 40$ ); when the ratio is in the range of 90:100 to 40:100, the optical  
15 material satisfies the region ( $n_d \leq -0.01v_d + 1.70$ ,  $v_d \leq 40$ ).

By mixing the low-index material ( $n_d \leq 1.40$ ) with the large-dispersion material ( $v_d \leq 15$ ) in this way, the optical material can be obtained in the region ( $n_d \leq$   
20  $-6.667 \times 10^{-3}v_d + 1.70$ ,  $v_d \leq 40$ ) or in the region ( $n_d \leq -0.01v_d + 1.70$ ,  $v_d \leq 40$ ).

Among the materials such as  $\text{TiO}_2$  ( $n_d$ ,  $v_d = 2.2652$ , 11.8),  $\text{Nb}_2\text{O}_5$  ( $n_d$ ,  $v_d = 2.367$ , 14.0), ITO ( $n_d$ ,  $v_d = 1.8581$ , 5.53),  $\text{Cr}_2\text{O}_3$  ( $n_d$ ,  $v_d = 2.2178$ , 13.4),  $\text{BaTiO}_3$  ( $n_d$ ,  $v_d =$   
25 2.4362, 11.3), etc. being large-dispersion materials, ITO ( $n_d$ ,  $v_d = 1.8581$ , 5.53) has the Abbe's number ( $v_d$ ) of half or less those of the other high-dispersion

materials and thus demonstrates extremely high dispersion.

For that reason, as to ITO ( $n_d$ ,  $v_d = 1.8581, 5.53$ ), even if the low-index material is a material having the index of  $n_d \geq 1.45$  but  $n_d \leq 1.55$ , the resultant optical material satisfies the region of ( $n_d \leq -6.667 \times 10^{-3}v_d + 1.70$ ,  $v_d \leq 40$ ).

The mixing material as the substance of  $n_d \leq 1.55$  can be selected from the general-purpose optical materials as typified by polymethyl methacrylate (hereinafter referred to as PMMA;  $n_d = 1.48$ ), amorphous polyolefin (hereinafter referred to as APO;  $n_d = 1.525$ ), some of copolymers of methyl methacrylate and styrene (hereinafter referred to as MS;  $n_d = 1.53$  to  $1.571$ ), and so on.

Since ITO is an electrically conductive substance, it can also reduce adhesion of dust to the lens surface due to charging specific to the optical resins for optical use.

Since ITO shows absorption in the ultraviolet region, it acts as a UV absorber and thus relieves deterioration of the resin due to ultraviolet rays.

The optical material in the region of ( $n_d \leq -6.667 \times 10^{-3}v_d + 1.70$ ,  $v_d \leq 40$ ) can be realized by using the material of  $1.45 \leq n_d \leq 1.55$  as the low-index material and using the particles having  $v_d \leq 10$  and the particle size of 2 to 100 nm as the large-dispersion material.

There are no specific restrictions on the low-index material for realizing this optical material, but it can be one of the general-purpose optical resins such as PMMA ( $n_d = 1.48$ ), APO ( $n_d = 1.525$ ), some of MS ( $n_d = 1.533$ ; the refractive index differs depending upon the copolymerization ratio of methyl methacrylate and styrene monomers), and so on.

The high-dispersion material for realizing the above optical material is desirably ITO ( $n_d, v_d = 1.8581, 5.53$ ). The reason is that there is no other material having the Abbe's number of not more than 10 in the visible region (light of 400 to 700 nm) than ITO ( $n_d, v_d = 1.8581, 5.53$ ).

There are no specific restrictions on the particle size of the particles of ITO ( $n_d, v_d = 1.8581, 5.53$ ) as long as it does not affect the scattering of light. However, the particle size is desirably not more than 100 nm.

For uniformly dispersing the above ITO particles, for example, in PMMA ( $n_d = 1.48$ ), APO ( $n_d = 1.525$ ), and some of MS ( $n_d = 1.533$ ) described above, it is effective to modify the surface of the ITO particles with an alkyl group having the nature of being compatible with PMMA.

A low-index high-dispersion optical material satisfying the region ( $n_d \leq -6.667 \times 10^{-3}v_d + 1.70$ ,  $v_d \leq 40$ ) can be obtained in either of the following cases:

where the mixing ratio of PMMA and ITO ( $n_d$ ,  $v_d = 1.8581$ , 5.53) is in the range of 30:100 to 250:100; where the mixing ratio of APO ( $n_d = 1.525$ ) and ITO ( $n_d$ ,  $v_d = 1.8581$ , 5.53) is in the range of 44:100 to 150:100;

5 where the mixing ratio of MS ( $n_d = 1.533$ ) and ITO ( $n_d$ ,  $v_d = 1.8581$ , 5.53) is in the range of 43:100 to 140:100.

Since ITO is the electroconductive material, the chargeability of the above optical materials is also reduced.

10 The aforementioned material of 2-ethoxy-ethyl methacrylate is the material realizing ( $n_d = 1.483$ ,  $v_d = 32$ ) by decreasing the density in terms of the structure, but the structure thereof is different from the structure of the present optical materials in the

15 range ( $n_d \leq -6.667 \times 10^{-3}v_d + 1.70$ ,  $v_d \leq 40$ ) formed by mixing the low-index material ( $n_d \leq 1.55$ ) with the large-dispersion material ( $v_d \leq 10$ ). In addition, the method with 2-ethoxy-ethyl methacrylate provides less freedom for adjusting the levels of refractive index

20 and index dispersion than the method of the present embodiment.

Specific examples of the optical materials according to the present invention will be described below.

25 <Example 1>

The particles of  $\text{Si}_x\text{-Ti}_{(1-x)}\text{O}_2$ , the surface of which was modified with a fluorine surfactant according to



Using measurements of the reflectance and transmittance of the above mixture films, absorption-scattering (rate) was calculated according to the equation below.

5           absorption-scattering (rate) = 1 - (two-side reflectance) - (transmittance).

The results of the refractive index and dispersion are presented in Table 1 and Fig. 9.

10           The mixtures at the ratios of  $\text{Si}_x\text{-Ti}_{(1-x)}\text{O}_2$  : amorphous fluororesin = 18.3:100 and 41.1:100 had the refractive index and dispersion of  $(n_d, v_d) = (1.337, 53.9)$  and  $(1.383, 41.0)$ , respectively, and were off the region of  $n_d \leq -6.667 \times 10^{-3}v_d + 1.70$ ,  $v_d \leq 40$ . However, since the absorption-scattering (rate) of the mixtures is not more than 0.01, they can be used as optical films without problems.

15           The mixture at the ratio of  $\text{Si}_x\text{-Ti}_{(1-x)}\text{O}_2$  : amorphous fluororesin = 70.5:100 had the refractive index and dispersion of  $(n_d, v_d) = (1.427, 34.9)$ , and was present within the region of  $n_d \leq -6.667 \times 10^{-3}v_d + 1.70$ ,  $v_d \leq 40$ . The absorption-scattering (rate) thereof was not more than 0.01, and the mixture can be used as an optical film without problems.

20           The mixture at the ratio of  $\text{Si}_x\text{-Ti}_{(1-x)}\text{O}_2$  : amorphous fluororesin = 88.5:100 showed great scattering with the absorption-scattering (rate) of 0.04 and the refractive index and index dispersion thereof were unmeasurable.

[Method 1] Preparation of solution in which particles of  $\text{Si}_x\text{-Ti}_{(1-x)}\text{O}_2$  modified with fluorine base surfactant were dispersed

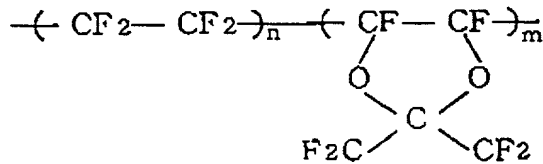
4 g of a fluorine base surfactant, N-(3-  
5 (trimethoxysilyl) propyl)-N-propylperfluorooctanesulfonamide, was added to a solution containing 20 g of particles of  $\text{Si}_x\text{-Ti}_{(1-x)}\text{O}_2$  (5 to 20 nm) in 500 g of 2, 2, 3, 3, 3-pentafluoro-1-propanol, and thereafter the mixture was put into 0.21  
10 ml of 1N hydrochloric acid. The mixture was agitated at 25°C for 24 hours to modify the surface of the particles. After that, 250 g of the fluorine solvent [the molecular structure of  $(\text{C}_4\text{F}_9)_3\text{N}$ ] was added to the mixture and fractional distillation was conducted at  
15 90°C to remove the pentafluoro propanol and isopropanol etc. made by hydrolysis, thereby obtaining the fluorine solvent solution in which the particles of  $\text{Si}_x\text{-Ti}_{(1-x)}\text{O}_2$  were dispersed.

<Comparative Example 1>

20 In contrast to Example 1, the particles of  $\text{Si}_x\text{-Ti}_{(1-x)}\text{O}_2$  with the refractive index and dispersion of  $(n_d, v_d) = (1.636, 29.1)$  were used and the index dispersion of the high-dispersion material was  $v_d > 25$ .

The particles of  $\text{Si}_x\text{-Ti}_{(1-x)}\text{O}_2$ , the surface of which  
25 was modified with the fluorine surfactant according to [Method 1] described above and the refractive index and the index dispersion of which were adjusted to  $(n_d, v_d)$

= (1.636, 29.1), and the amorphous fluororesin ( $n_d = 1.29$ ) represented by the chemical formula below, were mixed at the weight ratios of  $\text{Si}_x\text{-Ti}_{(1-x)}\text{O}_2$  : amorphous fluororesin = 17.3:100, 38.9:100, 66.7:100, and 83.8:100 in the fluorine solvent [the molecular structure of  $(\text{C}_4\text{F}_9)_3\text{N}$ ], which was a solvent in which the amorphous fluororesin was soluble.



The percentage of the amorphous fluororesin to the solvent was 4%. A coating of each of the above solutions was formed on the substrate of BK7 of  $\phi 49 \times 2\text{t}$  by the dip coat method and was heated in the furnace at  $180^\circ\text{C}$  for 20 minutes to obtain a film of the mixture of the amorphous fluororesin and the particles of  $\text{Si}_x\text{-Ti}_{(1-x)}\text{O}_2$ .

The refractive index and dispersion of the above mixture films were measured by the spectral ellipsometer (VASE: available from J. A. Woollam. Co., Inc.).

The two-side reflectance and transmittance of the above mixture films were measured by the spectrophotometer (U4000: available from Hitachi, Ltd.).

Using measurements of the reflectance and

transmittance of the above mixture films, the absorption-scattering (rate) was calculated according to the equation below.

absorption-scattering (rate) = 1 - (two-side  
5 reflectance) - (transmittance).

The results of the refractive index and dispersion are presented in Table 1 and Fig. 9.

The mixture at the ratio of  $\text{Si}_x\text{-Ti}_{(1-x)}\text{O}_2$  : amorphous  
fluororesin = 83.8:100 demonstrated large scattering  
10 with the absorption-scattering (rate) of 0.04 and the refractive index and index dispersion thereof were unmeasurable.

The refractive index and index dispersion of the mixtures at the ratios of  $\text{Si}_x\text{-Ti}_{(1-x)}\text{O}_2$  : amorphous  
15 fluororesin = 17.3:100, 38.9:100, and 66.7:100 were  $(n_d, v_d) = (1.329, 62.1), (1.366, 48.9), \text{ and } (1.403, 42.1),$  respectively, and none of them existed in the region of  $n_d \leq -6.667 \times 10^{-3}v_d + 1.70, v_d \leq 40.$

<Example 2>

20 The present example was different from Example 1 in that the  $\text{TiO}_2$  particles with the refractive index and dispersion of  $(n_d, v_d) = (2.2652, 11.8)$  were used instead of the particles of  $\text{Si}_x\text{-Ti}_{(1-x)}\text{O}_2.$

The particles of  $\text{TiO}_2$ , the surface of which was  
25 modified with the fluorine surfactant according to [Method 2] described hereinafter, and the amorphous fluororesin ( $n_d = 1.29$ ) represented by the chemical



absorption-scattering (rate) = 1 - (two-side reflectance) - (transmittance).

The results of the refractive index and dispersion are presented in Table 2 and Fig. 10.

5       The mixture at the ratio of  $\text{TiO}_2$  : amorphous fluororesin = 4.5:100 had the refractive index and dispersion of  $(n_d, v_d) = (1.317, 48.3)$ , and was off the region of  $n_d \leq -6.667 \times 10^{-3}v_d + 1.70$ ,  $v_d \leq 40$ . The mixture had the absorption-scattering (rate) of not  
10 more than 0.01 and thus can be used as an optical film without problems.

      The mixtures at the ratios of  $\text{TiO}_2$  : amorphous fluororesin = 9.3:100, 19:100, 39:100, and 74:100 had the refractive index and dispersion of  $(n_d, v_d) =$   
15  $(1.343, 34.2)$ ,  $(1.394, 23.8)$ ,  $(1.478, 17.8)$ , and  $(1.591, 14.8)$ , respectively, and were within the region of  $n_d \leq -6.667 \times 10^{-3}v_d + 1.70$ ,  $v_d \leq 40$ . They had the absorption-scattering (rates) of not more than 0.01 and can be used as optical films without problems.

20       The mixture at the ratio of  $\text{TiO}_2$  : amorphous fluororesin = 120:100 demonstrated great scattering with the absorption-scattering (rate) of 0.06 and the refractive index and dispersion were unmeasurable.

      In this Example 2, when compared with Example 1,  
25 the region of  $n_d \leq -6.667 \times 10^{-3}v_d + 1.70$ ,  $v_d \leq 40$  can be realized in the range of weight ratios of  $(\text{TiO}_2$  : amorphous fluororesin =) 7:100 to 90:100 and, further,

the region of  $n_d \leq -0.01 \times 10^{-3}v_d + 1.70$ ,  $v_d \leq 40$  is also realized thereby.

[Method 2] Preparation of solution in which particles of  $\text{TiO}_2$  modified with fluorine base surfactant were dispersed

4 g of the fluorine base surfactant, N-(3-(trimethoxysilyl) propyl)-N-propylperfluorooctanesulfonamide, was added to a solution containing 20 g of  $\text{TiO}_2$  particles (5 to 20 nm) in 500 g of 2, 2, 3, 3, 3-pentafluoro-1-propanol and thereafter 0.21 ml of 1N hydrochloric acid was added thereto. The mixture was stirred at 25°C for 24 hours to modify the surface of the particles. After that, 250 g of the fluorine solvent [the molecular structure of  $(\text{C}_6\text{F}_5)_3\text{N}$ ] was added thereto and fractional distillation was conducted at 90°C to remove the pentafluoro propanol, and isopropanol etc. created by hydrolysis, thereby preparing the fluorine solvent solution in which the  $\text{TiO}_2$  particles were dispersed.

<Comparative Example 2>

Comparative Example 2 was different from Example 2 in that a polymer with the refractive index ( $n_d = 1.52$ ) was used instead of the amorphous fluororesin ( $n_d = 1.29$ ).

The  $\text{TiO}_2$  particles and PVA ( $n_d = 1.52$ ) were blended at the weight ratios of  $\text{TiO}_2$  : PVA = 2 to 200:100 in a solution (water : MeOH = 1:1). The percentage of PVA

to the solvent was 4%. A coating of each of the above solutions was formed on the substrate of BK7 of  $\phi 49 \times 2t$  by the dip coat method and thereafter it was heated in the furnace at  $180^{\circ}\text{C}$  for 20 minutes, thereby obtaining a mixture film of PVA and the  $\text{TiO}_2$  particles.

The refractive index and dispersion of the above mixture films were measured by the spectral ellipsometer (VASE: available from J. A. Woollam. Co., Inc.).

The two-side reflectance and transmittance of the above mixture films were measured by the spectrophotometer (U4000: available from Hitachi, Ltd.).

Using measurements of the reflectance and transmittance of the above mixture films, the absorption-scattering (rate) was calculated according to the equation below.

absorption-scattering (rate) =  $1 - (\text{two-side reflectance}) - (\text{transmittance})$ .

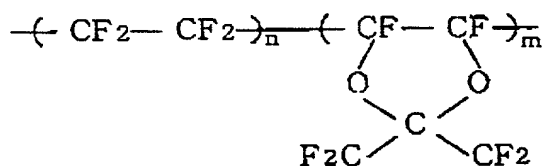
The results of the refractive index and dispersion are presented in Table 3 and Fig. 11.

The refractive index and dispersion of the mixtures were  $(n_d, v_d) = (1.564, 33.0), (1.590, 23.9), (1.633, 18.1), \text{ and } (1.691, 15.2)$ , neither of which was present in the region of  $n_d \leq -6.667 \times 10^{-3}v_d + 1.70, v_d \leq 40$ .

<Example 3>

The optical material adjusted to  $(n_d, v_d) = (1.523, 16.4)$  by mixing the amorphous fluororesin ( $n_d = 1.29$ ) represented by the chemical formula below and the  $\text{TiO}_2$  particles at the weight ratio of  $\text{TiO}_2$  : amorphous fluororesin = 51:100 as described in Example 2 was combined with PSK50:  $(n_d, v_d) = (1.5938, 60.9)$  available from Sumita Kogaku Kogyo, in the structure illustrated in Fig. 7.

10



15

The height  $d$  of the grating was  $8 \mu\text{m}$  and the diffraction efficiency was not less than 98% for the light at the wavelengths in the visible region of 400 to 700 nm. The height  $d$  of the grating is half that in Comparative Example 3 and this is advantageous in terms of forming of lens.

<Comparative Example 3>

20

p-methoxybenzyl methacrylate:  $(n_d, v_d) = (1.552, 32.5)$ , which was a material used heretofore, was combined with PSK50:  $(n_d, v_d) = (1.5938, 60.9)$ . The height  $d$  of the grating was  $15 \mu\text{m}$  and the diffraction efficiency was not less than 98% for the light at the wavelengths in the visible region of 400 to 700 nm.

25

<Example 4>

Particles of ITO ( $n_d, v_d = 1.8581, 5.53$ ), the

surface of which was modified with an alkyl group for enhancing the compatibility with PMMA according to [Method 3] described hereinafter, and PMMA ( $n_d = 1.48$ ) were mixed at the weight ratios of ITO : PMMA = 20:100, 30:100, 50:100, 120:100, 200:100, and 300:100 in methyl ethyl ketone (hereinafter referred to as MEK), which was a solvent in which PMMA was soluble. The percentage of PMMA to the solvent was 4%. A coating of each of the above solutions was formed on the substrate of BK7 of  $\phi 49 \times 2t$  by the dip coat method and thereafter heated in the furnace at 80°C for 20 minutes, thereby obtaining a mixture film of PMMA and ITO particles.

The refractive index and dispersion of the above mixture films were measured by the spectral ellipsometer (VASE: available from J. A. Woollam. Co., Inc.).

The two-side reflectance and transmittance of the above mixture films were measured by the spectrophotometer (U4000: available from Hitachi, Ltd.).

Using measurements of the reflectance and transmittance of the above mixture films, the absorption-scattering (rate) was calculated according to the equation below.

absorption-scattering (rate) =  $1 - (\text{two-side reflectance}) - (\text{transmittance})$ .

The results of the refractive index and dispersion are presented in Table 4 and Fig. 12.

The mixture at the weight ratio of ITO : PMMA = 20:100 demonstrated the refractive index and dispersion of  $(n_d, v_d) = (1.507, 32)$ , and was off the region of  $n_d \leq -6.667 \times 10^{-3}v_d + 1.70, v_d \leq 40$ .

The mixture had the absorption-scattering (rate) of not more than 0.01 and thus can be used as an optical film without problems.

The mixtures at the weight ratios of ITO : PMMA = 30:100, 50:100, 120:100, and 200:100 had the refractive index and dispersion of  $(n_d, v_d) = (1.515, 26.6)$ ,  $(1.531, 20.1)$ ,  $(1.571, 13.2)$ , and  $(1.609, 10.2)$ , respectively, and were in the region of  $n_d \leq -6.667 \times 10^{-3}v_d + 1.70, v_d \leq 40$ .

The mixtures had the absorption-scattering (rates) of not more than 0.01 and thus can be used as optical films without problems.

The mixture at the weight ratio of ITO : PMMA = 300:100 had the refractive index and dispersion of  $(n_d, v_d) = (1.647, 8.6)$ , and was off the region of  $n_d \leq -6.667 \times 10^{-3}v_d + 1.70, v_d \leq 40$ .

The chargeability was measured with an electrostatic voltmeter after each sample was wiped twenty times with wiping fabric (Savina Minimax: available from Kanebo Gosen K.K.). PMMA alone demonstrated the chargeability of 3 kV but the mixtures

all showed no charge.

[Method 3] Preparation of solution in which the ITO particles modified with alkyl group were dispersed

4 g of  $\gamma$ -methacryloxypropyltrimethoxysilane was added into a solution containing 20 g of ITO particles (5 to 20 nm) in 500 g of MEK and thereafter 0.2 ml of 1N hydrochloric acid was added thereto. The mixture was stirred at 25°C for 24 hours to modify the surface of the particles.

10 <Comparative Example 4>

Comparative Example 4 was different from Example 4 in that the particles of  $\text{TiO}_2$  ( $n_d$ ,  $v_d = 2.2652$ , 11.8) were used and the index dispersion of the high-dispersion material was  $v_d > 10$ .

15 The  $\text{TiO}_2$  particles ( $n_d = 1.29$ ), the surface of which was modified with an alkyl group for enhancing the compatibility with PMMA according to [Method 4] described hereinafter, and PMMA were mixed at the weight ratios of  $\text{TiO}_2$  : PMMA = 20:100, 50:100, 85:100, 20 and 160:100 in MEK, which was a solvent in which PMMA was soluble. The percentage of PMMA to the solvent was 4%. A coating of each of the above solutions was formed on the substrate of BK7 of  $\phi 49 \times 2t$  by the dip coat method and thereafter was heated in the furnace at 25 80°C for 20 minutes, thereby obtaining a mixture film of PMMA and  $\text{TiO}_2$  particles.

The two-side reflectance and transmittance of the

above mixture films were measured by the spectrophotometer (U4000: available from Hitachi, Ltd.).

5 Using measurements of the reflectance and transmittance of the above mixture films, the absorption-scattering (rate) was calculated according to the equation below.

absorption-scattering (rate) =  $1 - (\text{two-side reflectance}) - (\text{transmittance})$ .

10 The results of the refractive index and dispersion are presented in Table 4 and Fig. 12.

The mixtures at the weight ratios of  $\text{TiO}_2$  : PMMA = 20:100, 50:100, 85:100, and 160:100 had the refractive index and dispersion of  $(n_d, v_d) = (1.519, 40.2)$ ,  
15  $(1.585, 26)$ ,  $(1.63, 21.7)$ , and  $(1.717, 17.5)$ , respectively, and were off the region of  $n_d \leq -6.667 \times 10^{-3}v_d + 1.70$ ,  $v_d \leq 40$ .

The chargeability was measured with the electrostatic voltmeter after each sample was wiped  
20 twenty times with the wiping fabric (Savina Minimax: available from Kanebo Gosen K.K.). PMMA alone demonstrated the chargeability of 3 kV. The mixtures showed the chargeability of 1 kV to 2 kV.

[Method 4] Preparation of solution in which  $\text{TiO}_2$   
25 particles modified with alkyl group were dispersed

4 g of  $\gamma$ -methacryloxypropyltrimethoxysilane was added to a solution containing 20 g of  $\text{TiO}_2$  particles (5

to 20 nm) in 500 g of MEK, and thereafter 0.2 ml of 1N hydrochloric acid was added thereto. The mixture was stirred at 25°C for 24 hours to modify the surface of the particles.

5 <Comparative Example 5>

Comparative Example 5 was different from Example 4 in that polycarbonate (hereinafter referred to as PC) with the refractive index ( $n_d = 1.59$ ) was used instead of PMMA ( $n_d = 1.48$ ).

10 The ITO particles and PC ( $n_d = 1.59$ ) were mixed at the weight ratios of ITO : PC = 20:100, 50:100, 85:100, and 160:100 in dichloroethane. The percentage of the solute to the solvent of dichloroethane was 4%. A coating of each of the above solutions was formed on  
15 the substrate of BK7 of  $\phi 49 \times 2t$  by the dip coat method and thereafter was heated in the furnace at 110°C for 20 minutes, thereby obtaining a mixture film of PC and ITO particles.

The refractive index and dispersion of the above  
20 mixture films were measured by the spectral ellipsometer (VASE: available from J. A. Woollam. Co., Inc.).

The two-side reflectance and transmittance of the above mixture films were measured by the  
25 spectrophotometer (U4000: available from Hitachi, Ltd.).

Using measurements of the reflectance and

transmittance of the above mixture films, the absorption-scattering (rate) was calculated according to the equation below.

absorption-scattering (rate) = 1 - (two-side  
5 reflectance) - (transmittance).

The results of the refractive index and dispersion are presented in Table 4 and Fig. 12.

The mixtures had the refractive index and dispersion of  $(n_d, v_d) = (1.596, 23.2), (1.614, 17.5),$   
10  $(1.628, 14.7),$  and  $(1.657, 11.4),$  respectively, and all were off the region of  $n_d \leq -6.667 \times 10^{-3}v_d + 1.70,$   
 $v_d \leq 40.$

The chargeability was measured with the electrostatic voltmeter after each sample was wiped  
15 twenty times with the wiping fabric (Savina Minimax: available from Kanebo Gosen K.K.). PC alone demonstrated the chargeability of 3 kV, but the mixtures all showed no charge.

<Example 5>

20 The particles of ITO ( $n_d, v_d = 1.8581, 5.53$ ), the surface of which was modified with an alkyl group for enhancing the compatibility with APO ( $n_d = 1.525$ ) according to [Method 5] described hereinafter, and APO  
25 ( $n_d = 1.525$ ) were mixed at the weight ratios of ITO : APO = 25:100, 45:100, 70:100, 100:100, 135:100, and 220:100 in xylene, which was a solvent in which APO was soluble. The percentage of APO to the solvent was 4%.

A coating of each of the above solutions was formed on the substrate of BK7 of  $\phi 49 \times 2t$  by the dip coat method and thereafter was heated in the furnace at  $110^\circ\text{C}$  for 20 minutes, thereby obtaining a mixture film of APO and ITO particles.

The refractive index and dispersion of the above mixture films were measured by the spectral ellipsometer (VASE: available from J. A. Woollam. Co., Inc.).

The two-side reflectance and transmittance of the above mixture films were measured by the spectrophotometer (U4000: available from Hitachi, Ltd.).

Using measurements of the reflectance and transmittance of the above mixture films, the absorption-scattering (rate) was calculated according to the equation below.

$$\text{absorption-scattering (rate)} = 1 - (\text{two-side reflectance}) - (\text{transmittance}).$$

The results of the refractive index and dispersion are presented in Table 5 and Fig. 13.

The mixture at the weight ratio of ITO : APO = 25:100 had the refractive index and dispersion of  $(n_d, v_d) = (1.548, 26.6)$ , and was off the region of  $n_d \leq -6.667 \times 10^{-3}v_d + 1.70$ ,  $v_d \leq 40$ .

The mixture had the absorption-scattering (rate) of not more than 0.01 and can be used as an optical

film without problems.

The mixtures at the weight ratios of ITO : APO = 45:100, 70:100, 100:100, and 135:100 had the refractive index and dispersion of  $(n_d, v_d) = (1.562, 20.5)$ ,  
5  $(1.580, 16.2)$ ,  $(1.598, 13.6)$ , and  $(1.615, 11.8)$ , respectively, and they were in the region of  $n_d \leq -6.667 \times 10^{-3}v_d + 1.70$ ,  $v_d \leq 40$ .

The mixtures had the absorption-scattering (rates) of not more than 0.01 and can be used as optical films  
10 without problems.

The mixture at the weight ratio of ITO : APO = 220:100 had the refractive index and dispersion of  $(n_d, v_d) = (1.650, 9.63)$ , and was off the region of  $n_d \leq -6.667 \times 10^{-3}v_d + 1.70$ ,  $v_d \leq 40$ .

15 The chargeability was measured with the electrostatic voltmeter after each sample was wiped twenty times with the wiping fabric (Savina Minimax: available from Kanebo Gosen K.K.). APO alone demonstrated the chargeability of 3 kV, but the  
20 mixtures all showed no charge.

[Method 5] Preparation of solution in which ITO particles modified with alkyl group were dispersed

4 g of  $\beta$ -(3, 4 epoxycyclohexyl) ethyltrimethoxysilane was added to a solution  
25 containing 20 g of ITO particles (5 to 20 nm) in 500 g of n-butanol, and thereafter 0.2 ml of 1N hydrochloric acid was added thereto. The mixture was stirred at

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25°C for 24 hours to modify the surface of the particles.

After that, 250 g of xylene was added to the solution and n-butanol was removed by distillation to obtain a xylene solution of ITO particles.

<Example 6>

The surface of particles of ITO ( $n_d$ ,  $v_d$  = 1.8581, 5.53) was modified with an alkyl group for enhancing the compatibility with MS ( $n_d$  = 1.534) according to [Method 3].

The ITO particles and MS ( $n_d$  = 1.534) were mixed at the weight ratios of ITO : MS = 25:100, 45:100, 70:100, 100:100, 135:100, and 220:100 in MEK which was a solvent in which MS was soluble. The percentage of MS to the solvent was 4%. A coating of each of the above solutions was formed on the substrate of BK7 of  $\phi 49 \times 2t$  by the dip coat method and thereafter was heated in the furnace at 80°C for 20 minutes, thereby obtaining a mixture film of MS and ITO particles.

The refractive index and dispersion of the above mixture films were measured by the spectral ellipsometer (VASE: available from J. A. Woollam. Co., Inc.).

The two-side reflectance and transmittance of the above mixture films were measured by the spectrophotometer (U4000: available from Hitachi, Ltd.).



$$-6.667 \times 10^{-3}v_d + 1.70, v_d \leq 40.$$

The chargeability was measured with the electrostatic voltmeter after each sample was wiped twenty times with the wiping fabric (Savina Minimax: available from Kanebo Gosen K.K.). PC alone demonstrated the chargeability of 3 kV, but the mixtures all showed no charge.

<Example 7>

The optical material adjusted to  $(n_d, v_d) = (1.531, 20.1)$  by mixing PMMA and ITO particles at the weight ratio of ITO : PMMA = 50:100 as described in Example 4, was combined with PSK50:  $(n_d, v_d) = (1.5938, 60.9)$  available from Sumita Kogaku Kogyo, in the structure illustrated in Fig. 8. The height  $d$  of the grating was 9.98  $\mu\text{m}$  and the diffraction efficiency was not less than 98% for the light at the wavelengths in the visible region of 400 to 700 nm.

The height of the grating was approximately two thirds of that in Comparative Example 3 and this is advantageous in terms of forming of lens.

The chargeability was measured with the electrostatic voltmeter after the sample was wiped twenty times with the wiping fabric (Savina Minimax: available from Kanebo Gosen K.K.). The sample showed no charge.

<Comparative Example 6>

The conventional material of p-methoxybenzyl

methacrylate: ( $n_d$ ,  $v_d$ ) = (1.552, 32.5) was combined with  
PSK50: ( $n_d$ ,  $v_d$ ) = (1.5938, 60.9). The height  $d$  of the  
grating was 15  $\mu\text{m}$  and the diffraction efficiency was  
not less than 98% for the light at the wavelengths in  
the visible region of 400 to 700 nm.

The chargeability was measured with the  
electrostatic voltmeter after the sample was wiped  
twenty times with the wiping fabric (Savina Minimax:  
available from Kanebo Gosen K.K.). The sample showed  
the charge of 1 kV.

<Example 8>

Example 8 is different from Example 1 in that the  
 $\text{TiO}_2$  particles with the refractive index and dispersion  
of ( $n_d$ ,  $v_d$  = 2.2652, 11.8) were used instead of the  
particles of  $\text{Si}_x\text{-TiO}_2$  and the dimethylsilicone resin was  
used instead of the amorphous fluororesin.

The  $\text{TiO}_2$  particles, the surface of which was  
modified with a functional group of dimethylsilicone  
according to [Method 6] described hereinafter, and the  
elastomer of dimethylsilicone resin ( $n_d$  = 1.43) were  
mixed at the weight ratios of  $\text{TiO}_2$  : dimethylsilicone  
resin = 5:100, 10:100, 20:100, 38:100, 55:100, and  
88:100 in  $n$ -heptane, which was a solvent in which the  
elastomer of dimethylsilicone was soluble.

The percentage of the dimethylsilicone resin to  
the solvent was 4%. A coating of each of the above  
solutions was formed on the substrate of BK7 of  $\phi 49 \times$





were dispersed

4 g of the silicone base surfactant was added to a solution containing 20 g of  $\text{TiO}_2$  particles (5 to 20 nm) in 500 g of n-butanol, and thereafter 0.2 ml of 1N hydrochloric acid was added thereto. The solution was stirred at 25°C for 24 hours to modify the surface of the particles.

After that, 250 g of n-heptane was added to the solution and the fractional distillation was conducted at 100°C to remove n-butanol, and alcohols etc. created by hydrolysis, thereby preparing the n-heptane solution in which the  $\text{TiO}_2$  particles were dispersed.

The optical materials prepared according to the above method of the present embodiment are used for the optical systems such as image pickup optical systems, projection optical systems, illumination systems, and so on, or for the optical devices such as cameras, microscopes, binoculars, and so on.

Each of the above examples permits attainment of the optical materials satisfying the region ( $n_d \leq -6.667 \times 10^{-3}v_d + 1.70$ ,  $v_d \leq 40$ ) or  $n_d \leq -0.01v_d + 1.70$ ,  $v_d \leq 40$  and being indispensable for performance enhancement of the refracting optical systems and stack optical elements, and the optical systems using it.

Particularly, the optical material satisfying the low-index high-dispersion region ( $n_d \leq -6.667 \times 10^{-3}v_d + 1.70$ ,  $v_d \leq 40$ ) can be realized by mixing the low-index

material ( $n_d \leq 1.45$ ) with the large-dispersion material ( $v_d \leq 25$ ) as described above.

In that case, the optical material can be one better satisfying the region ( $n_d \leq -6.667 \times 10^{-3}v_d + 1.70$ ,  $v_d \leq 40$ ) where the particles (2 to 100 nm) of the composite metal oxide of titanium and silicon ( $\text{Si}_x\text{-Ti}_{(1-x)}\text{O}_2$ ) ( $v_d = 24.4$ ) are used as the large-dispersion material, the amorphous fluororesin ( $n_d = 1.29$  to  $1.38$ ) is used as the low-index material, and they are mixed in the weight ratio range of ( $\text{Si}_x\text{-Ti}_{(1-x)}\text{O}_2$  : fluororesin) of 45:100 to 75:100.

Further, the low-index high-dispersion region ( $n_d \leq -0.01v_d + 1.70$ ,  $v_d \leq 40$ ) can be realized by mixing the low-index material ( $n_d \leq 1.40$ ) with the particles of the large-dispersion material ( $v_d \leq 15$ ).

In that case, the low index and high dispersion tendency is enhanced more where the particles (2 to 100 nm) of  $\text{TiO}_2$  ( $n_d$ ,  $v_d = 2.2652$ ,  $11.8$ ) are used as the large-dispersion material, the amorphous fluororesin ( $n_d = 1.29$  to  $1.38$ ) as the low-index material, and they are mixed in the weight ratio range of ( $\text{TiO}_2$  : fluororesin) of 7:100 to 90:100.

This permits the supply of the optical materials in the region ( $n_d \leq -6.667 \times 10^{-3}v_d + 1.70$ ,  $v_d \leq 40$ ) and in the region ( $n_d \leq -0.01v_d + 1.70$ ,  $v_d \leq 40$ ) and thus facilitates achievement of higher performance of the optical systems such as the refracting optical systems,

the stack optical elements, and so on.

The low-index high-dispersion region ( $n_d \leq -6.667 \times 10^{-3}v_d + 1.70$ ,  $v_d \leq 40$ ) can also be realized by mixing the low-index material ( $1.45 \leq n_d \leq 1.55$ ) with the  
5 large-dispersion material ( $v_d \leq 10$ ) as described above.

In that case, the optical material can be one better satisfying the region ( $n_d \leq -6.667 \times 10^{-3}v_d + 1.70$ ,  $v_d \leq 40$ ) where the particles (2 to 100 nm) of ITO ( $v_d = 5.53$ ) are used as the large-dispersion material,  
10 either polymethyl methacrylate (PMMA,  $n_d = 1.48$ ), amorphous polyolefin (APO,  $n_d = 1.525$ ), or the copolymer of methyl methacrylate and styrene (MS,  $n_d = 1.533$ ) as the low-index material, and they are mixed in the weight ratio range of (ITO : PMMA) of 30:100 to  
15 250:100, in the weight ratio range of (ITO : APO) of 44:100 to 150:100, or in the weight ratio range of (ITO : MS) of 43:100 to 140:100.

This permits the supply of the optical materials in the region ( $n_d \leq -6.667 \times 10^{-3}v_d + 1.70$ ,  $v_d \leq 40$ ) by  
20 use of the materials of  $1.45 \leq n_d \leq 1.55$  such as PMMA ( $n_d = 1.48$ ), APO ( $n_d = 1.525$ ), MS ( $n_d = 1.533$ ), etc. which are commonly used as optical materials, and thus facilitates achievement of higher performance of the refracting optical systems and stack diffracting  
25 optical elements.

At the same time, since ITO is the electroconductive substance, it can reduce the



Table 1

	Refractive Index and Abbe's Number of SiO <sub>2</sub> - TiO <sub>2</sub> ( $n_d$ , $v_d$ )	Weight Ratio (Si <sub>x</sub> -Ti <sub>1-x</sub> O <sub>2</sub> : TefronAF)	Refractive Index $n_d$	Abbe's Number $v_d$	Absorption / Scattering ( $\lambda=500\text{nm}$ ) = 1 - (Transmittance) - (Two-side Reflectance)
Example 1	1.703, 24.4	18.3 : 100	1.337	53.9	less than 0.005
		41.1 : 100	1.383	41.0	less than 0.005
		70.5 : 100	1.427	34.9	less than 0.005
		88.5 : 100	----	----	0.04
Comparative Example 1	1.636, 29.1	17.3 : 100	1.329	62.1	less than 0.005
		38.9 : 100	1.366	48.9	less than 0.005
		66.7 : 100	1.403	42.1	less than 0.005
		83.8 : 100	----	----	0.04

Table 2

	Weight Ratio (TiO <sub>2</sub> : TefronAF)	Refractive Index $n_d$	Abbe's Number $v_d$	Absorption / Scattering ( $\lambda=500\text{nm}$ ) = 1 - (Transmittance) - (Two-side Reflectance)
Example 2	4.5 : 100	1.317	48.3	less than 0.005
	9.3 : 100	1.343	34.2	less than 0.005
	19 : 100	1.394	23.8	less than 0.005
	39 : 100	1.478	17.8	less than 0.005
	74 : 100	1.591	14.8	0.007
	120 : 100	----	----	0.06

Table 3

	Weight Ratio (TiO <sub>2</sub> : Polymer)	Refractive Index $n_d$	Abbe's Number $v_d$	Absorption / Scattering ( $\lambda=500\text{nm}$ ) = 1 - (Transmittance) - (Two-side Reflectance)
Comparative Example 2	14.8 : 100	1.564	33.0	less than 0.005
	29.6 : 100	1.590	23.9	less than 0.005
	59.3 : 100	1.633	18.1	less than 0.005
	103.7 : 100	1.691	15.2	0.007

Table 4

	High Disperse Material and Low Refraction Material, and their Index and Abbe's Number ( $n_d, v_d$ )	Weight Ratio (High Disperse Material : Low Refraction Material)	Refractive Index $n_d$	Abbe's Number $v_d$	Absorption / Scattering ( $\lambda=500\text{nm}$ ) = 1 - (Transmittance) - (Two-side Reflectance)
Example 4	ITO ( $n_d, v_d = 1.8581, 5.53$ )	20 : 100	1.5065	32.1	less than 0.005
		30 : 100	1.5146	26.6	less than 0.005
	PMMA ( $n_d, v_d = 1.490, 57.6$ )	50 : 100	1.5308	20.1	less than 0.005
		120 : 100	1.5705	13.2	less than 0.005
		200 : 100	1.6093	10.2	less than 0.005
		300 : 100	1.6471	8.65	0.01
Comparative Example 4	TiO <sub>2</sub> ( $N_d, v_d = 2.265, 11.8$ )	20 : 100	1.5190	40.2	less than 0.005
		50 : 100	1.5847	25.9	less than 0.005
	PMMA ( $N_d, v_d = 1.490, 57.6$ )	85 : 100	1.6300	21.7	less than 0.005
		160 : 100	1.7169	17.5	less than 0.005
Comparative Example 5	ITO ( $N_d, v_d = 1.8581, 5.53$ )	20 : 100	1.5961	23.2	less than 0.005
		50 : 100	1.6137	17.5	less than 0.005
	PC ( $N_d, v_d = 1.58, 30.5$ )	85 : 100	1.6282	14.7	less than 0.005
		160 : 100	1.6569	11.4	less than 0.005

Table 5

	High Disperse Material and Low Refraction Material, and their Index and Abbe's Number ( $n_d$ , $v_d$ )	Weight Ratio (High Disperse Material : Low Refraction Material)	Refractive Index $n_d$	Abbe's Number $v_d$	Absorption / Scattering ( $\lambda=500\text{nm}$ ) = 1 - (Transmittance) - (Two-side Reflectance)
Example 5	ITO ( $n_d$ , $v_d$ = 1.8581, 5.53)	25 : 100	1.548	26.6	less than 0.005
		45 : 100	1.562	20.5	less than 0.005
	APO ( $n_d$ , $v_d$ = 1.525, 52.1)	70 : 100	1.580	16.2	less than 0.005
		100 : 100	1.598	13.6	less than 0.005
		135 : 100	1.615	11.8	less than 0.005
		220 : 100	1.650	9.63	0.01
Example 6	ITO ( $n_d$ , $v_d$ = 1.8581, 5.53)	25 : 100	1.555	24.1	less than 0.005
		45 : 100	1.569	19.2	less than 0.005
	MS ( $n_d$ , $v_d$ = 1.534, 41.5)	70 : 100	1.587	15.5	less than 0.005
		100 : 100	1.604	13.2	less than 0.005
		135 : 100	1.621	11.6	less than 0.005
		220 : 100	1.655	9.51	0.01

Table 6

	Weight Ratio (TiO <sub>2</sub> : Dimethyl Silicone)	Refractive Index $n_d$	Abbe's Number $v_d$	Absorption / Scattering ( $\lambda=500\text{nm}$ ) = 1 - (Transmittance) - (Two-side Reflectance)
Example 8	5 : 100	1.440	44.6	less than 0.005
	10 : 100	1.450	39.9	less than 0.005
	20 : 100	1.471	33.5	less than 0.005
	38 : 100	1.502	27.7	less than 0.005
	55 : 100	1.533	24.2	less than 0.005
	88 : 100	1.582	20.6	0.02

WHAT IS CLAIMED IS: .

1. An optical material which is a mixture of materials comprising a first material having a refractive index of not more than 1.45 for the d-line and a second material having an Abbe's number of not more than 25, wherein a relation between a refractive index for the d-line ( $n_d$ ) and an Abbe's number ( $v_d$ ) is defined as follows:

$$n_d \leq -6.667 \times 10^{-3} v_d + 1.70.$$

2. The optical material of Claim 1 wherein said Abbe's number ( $v_d$ ) is less than 40.

3. The optical material of Claim 1 wherein said second material comprises particles having the grain size in the range of 2 to 100 nm.

4. The optical material of Claim 1 wherein said first material is an amorphous fluororesin.

5. The optical material of Claim 1 wherein said second material is particles of a composite metal oxide of titanium and silicon ( $\text{Si}_x\text{-Ti}_{(1-x)}\text{O}_2$ ) having the Abbe's number ( $v_d$ ) of 24.4.

6. The optical material of Claim 1 wherein said first material is an amorphous fluororesin, said second

material is particles of a composite metal oxide of titanium and silicon ( $\text{Si}_x\text{-Ti}_{(1-x)}\text{O}_2$ ) having the Abbe's number ( $v_d$ ) of 24.4, and a weight ratio of the particles and said amorphous fluororesin is in the range of 45:100 to 75:100.

7. The optical material of Claim 1 wherein said first material is a dimethylsilicone resin.

8. The optical material of Claim 1 wherein said second material comprises particles of titanium oxide ( $\text{TiO}_2$ ).

9. The optical material of Claim 1 wherein said first material is a dimethylsilicone resin, said second material is particles of titanium oxide ( $\text{TiO}_2$ ), and a weight ratio of the titanium oxide and said dimethylsilicone resin is in the range of 18:100 to 70:100.

10. An optical material wherein a relation between a refractive index for the d-line ( $n_d$ ) and an Abbe's number ( $v_d$ ) satisfies the following condition:

$$n_d \leq -0.01v_d + 1.70.$$

11. An optical material which is a mixture of materials comprising a first material having a

refractive index of not more than 1.40 for the d-line and a second material having an Abbe's number of not more than 15, wherein a relation between a refractive index for the d-line ( $n_d$ ) and an Abbe's number ( $v_d$ ) is defined as follows:

$$n_d \leq -0.01v_d + 1.70.$$

12. The optical material of Claim 10 or 11 wherein said Abbe's number ( $v_d$ ) is not more than 40.

13. The optical material of Claim 11 wherein said second material comprises particles having the grain size in the range of 2 to 100 nm.

14. The optical material of Claim 11 wherein said first material comprises an amorphous fluororesin.

15. The optical material of Claim 11 wherein said second material comprises particles of titanium oxide ( $\text{TiO}_2$ ).

16. The optical material of Claim 11 wherein said first material is an amorphous fluororesin, said second material is particles of titanium oxide ( $\text{TiO}_2$ ), and a weight ratio of the titanium oxide and said amorphous fluororesin is in the range of 7:100 to 90:100.

17. An optical material which is a mixture of materials comprising a first material having a refractive index for the d-line in the range of 1.45 to 1.55 both inclusive and a second material having an  
5 Abbe's number of not more than 10, wherein a relation between a refractive index for the d-line ( $n_d$ ) and an Abbe's number ( $v_d$ ) is defined as follows:

$$n_d \leq -6.667 \times 10^{-3} v_d + 1.70.$$

10 18. The optical material of Claim 17 wherein said Abbe's number ( $v_d$ ) is not more than 40.

15 19. The optical material of Claim 17 wherein said second material comprises particles having the grain size in the range of 2 to 100 nm.

20. The optical material of Claim 17 wherein said second material is ITO (indium-tin-oxide).

20 21. The optical material of Claim 17 wherein said first material is polymethyl methacrylate.

25 22. The optical material of Claim 17 wherein said first material is polymethyl methacrylate, said second material is particles of ITO (indium-tin-oxide), and a weight ratio of the particles and said polymethyl methacrylate is in the range of 30:100 to 250:100.

23. The optical material of Claim 17 wherein said first material is an amorphous polyolefin.

5 24. The optical material of Claim 17 wherein said first material is an amorphous polyolefin, said second material is particles of ITO (indium-tin-oxide), and a weight ratio of the particles and said amorphous polyolefin is in the range of 44:100 to 150:100.

10 25. The optical material of Claim 17 wherein said first material is a copolymer of methyl methacrylate and styrene.

15 26. The optical material of Claim 17 wherein said first material is a copolymer resin of methyl methacrylate and styrene, said second material is particles of ITO (indium-tin-oxide), and a weight ratio of the particles and said copolymer resin is in the range of 43:100 to 140:100.

20 27. An optical member comprising the optical material as set forth in either one of Claims 1 to 26.

25 28. An optical system comprising the optical member of Claim 27.

29. A diffracting optical element using the

optical material as set forth in either one of Claims 1 to 26.

30. An optical system comprising the diffracting  
5 optical element of Claim 29.

31. An optical device comprising the optical  
system of Claim 28 or 30.

10 32. A method for producing an optical material,  
comprising a step of decreasing a filling factor of a  
first material, and a step of filling gaps of the first  
material of the decreased filling factor with a second  
material having an Abbe's number different from that of  
15 the first material, thereby producing an optical  
material having a desired refractive index and an  
Abbe's number.

20 33. An optical member comprising the material  
produced by the production method as set forth in Claim  
32.

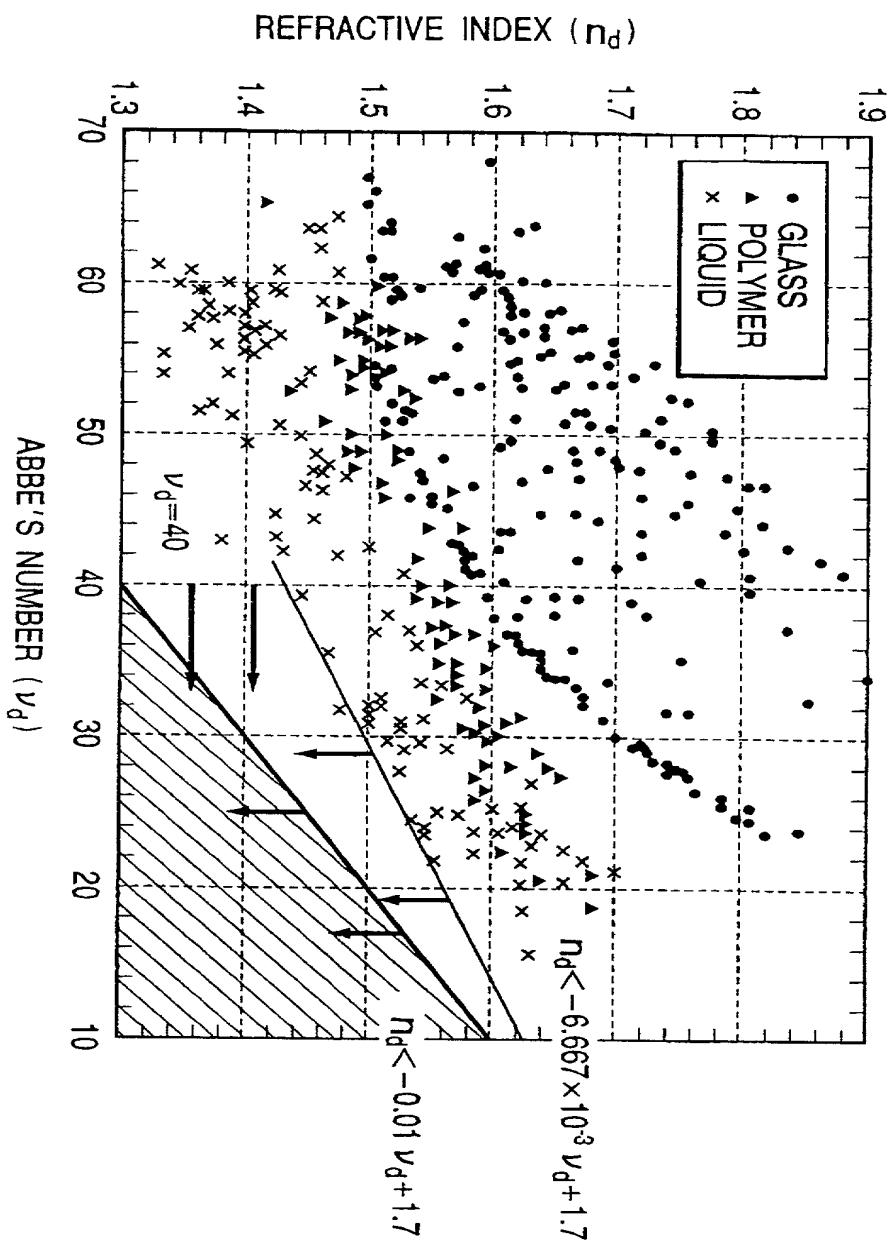
25 34. An optical system comprising the optical  
member of Claim 33.

35. The optical system of Claim 34 wherein said  
optical member is a diffracting optical element.





FIG. 1



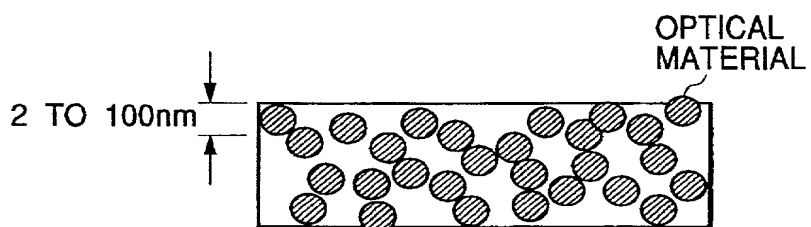
*FIG. 2*

FIG. 3

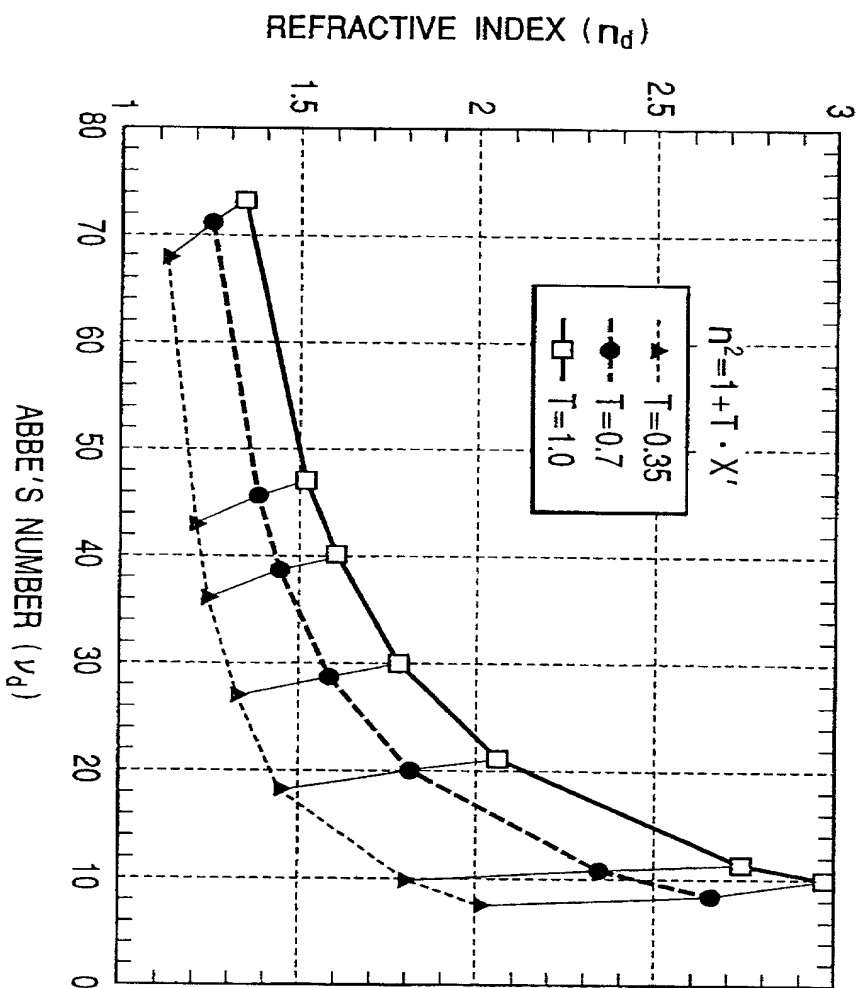


FIG. 4

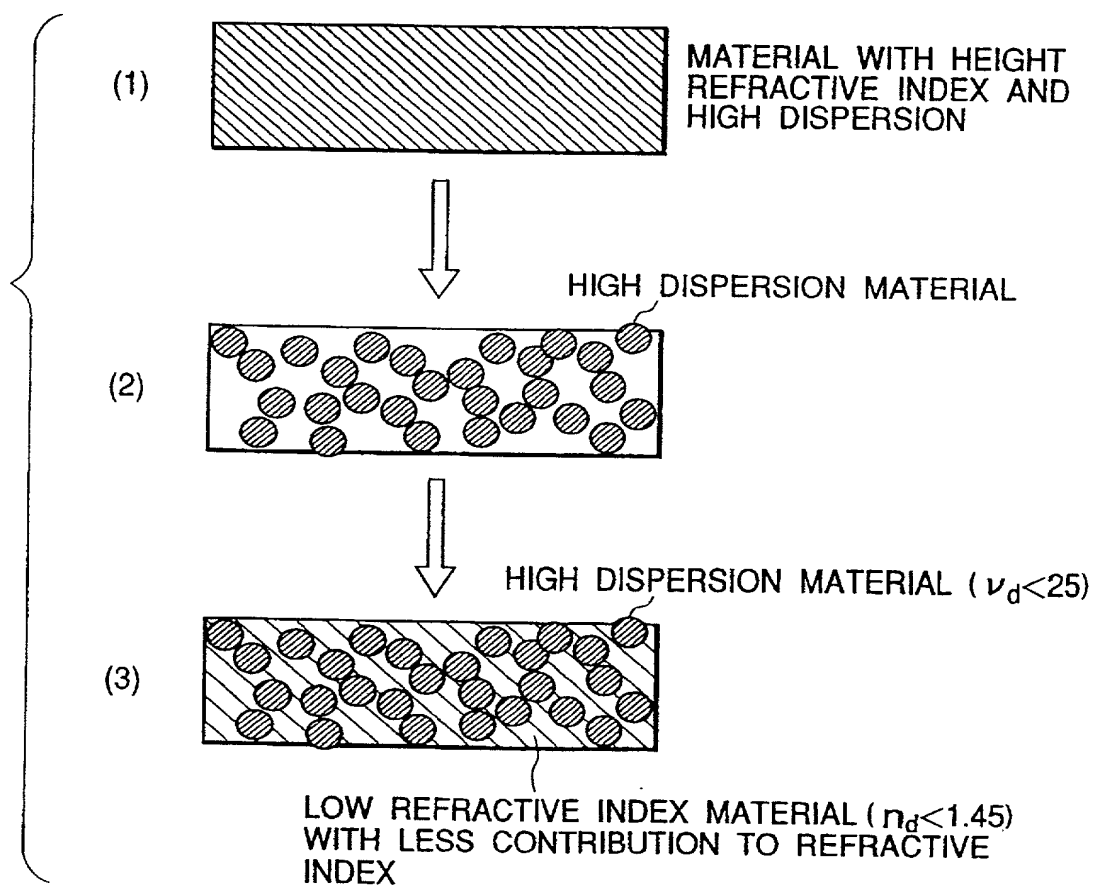
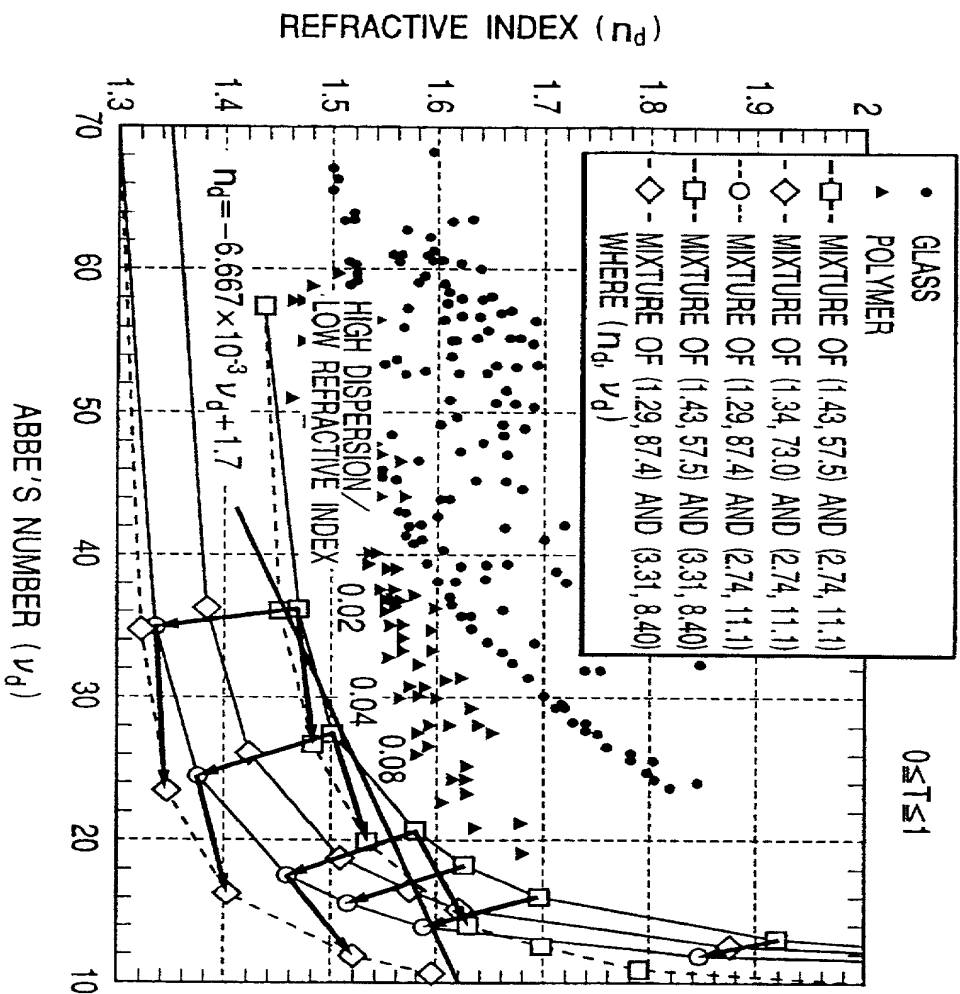
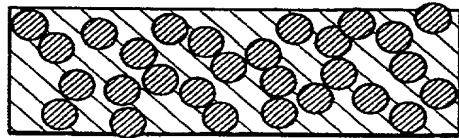


FIG. 5

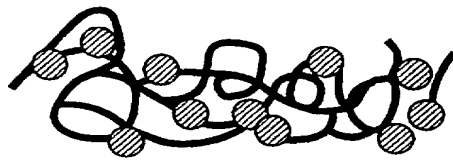
$$n^2 = 1 + T x_0 + (1 - T) x_1$$



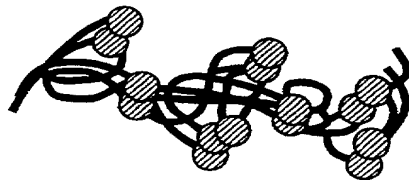
**FIG. 6**  
TYPE OF MIXTURE



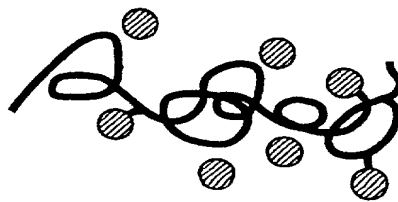
(1) NORMAL MIXTURE



(2) GUEST-HOST TYPE MIXTURE



(3) BACKBONE CHAIN TYPE MIXTURE



(4) SIDE CHAIN TYPE MIXTURE

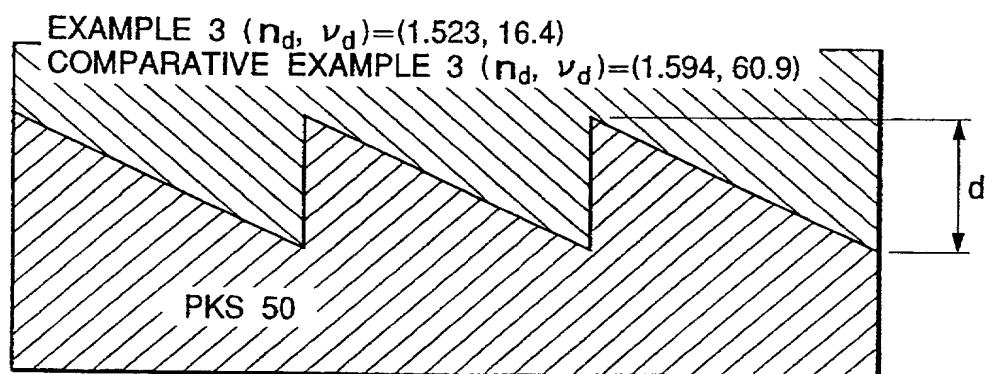
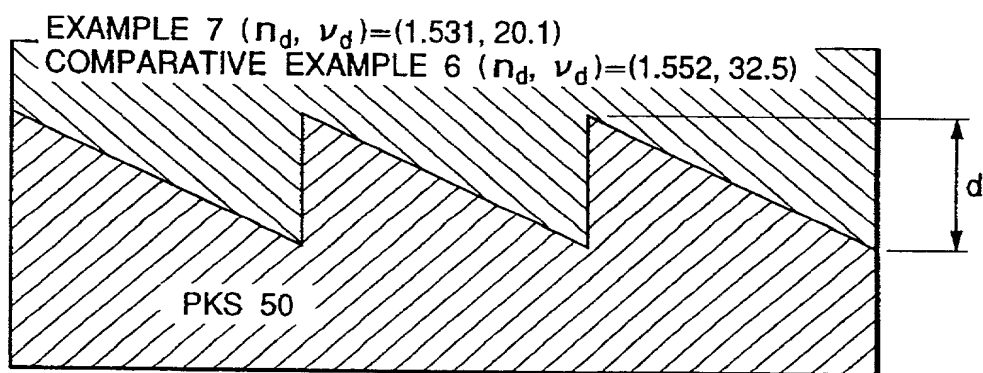
**FIG. 7****FIG. 8**

FIG. 9

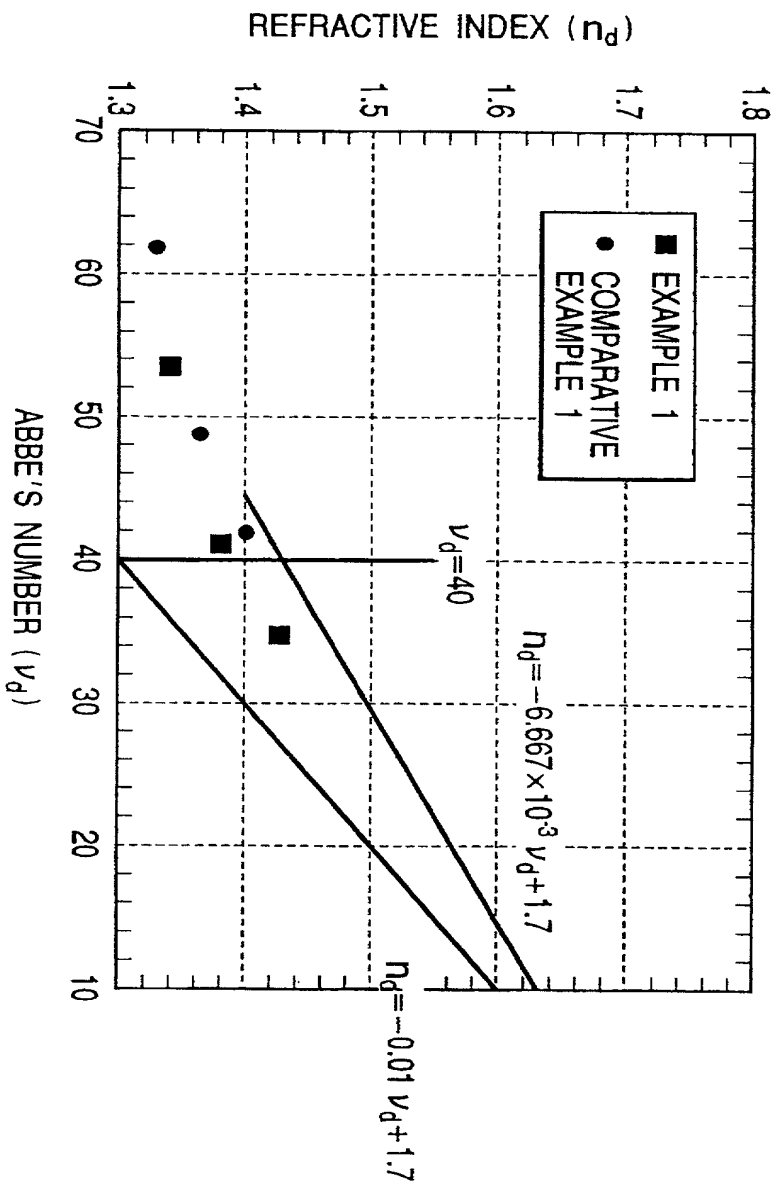


FIG. 10

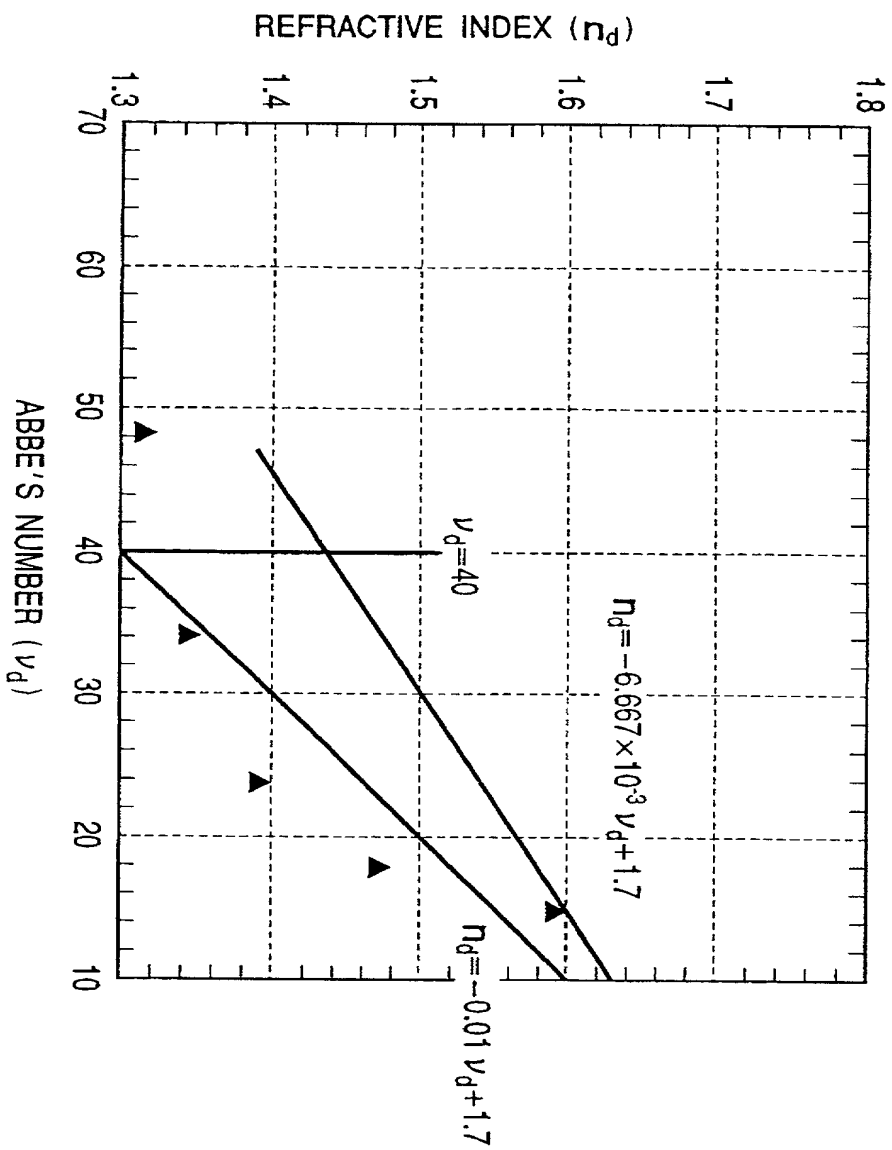


FIG. 11

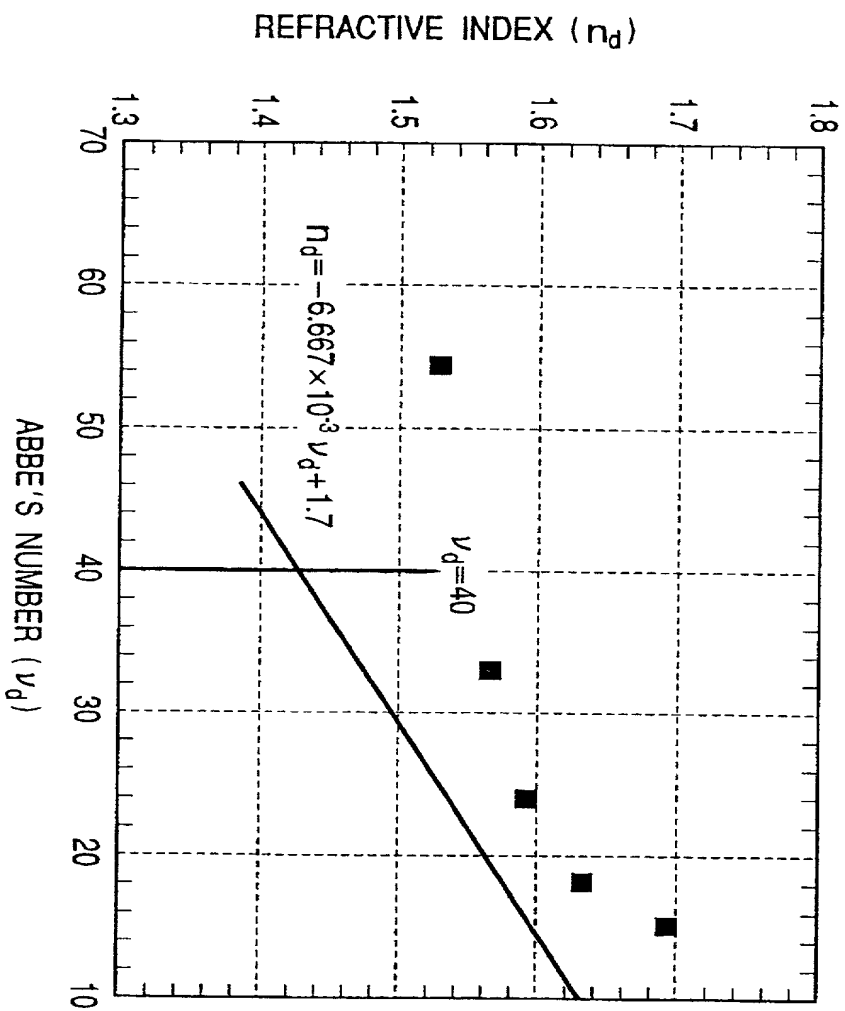


FIG. 12

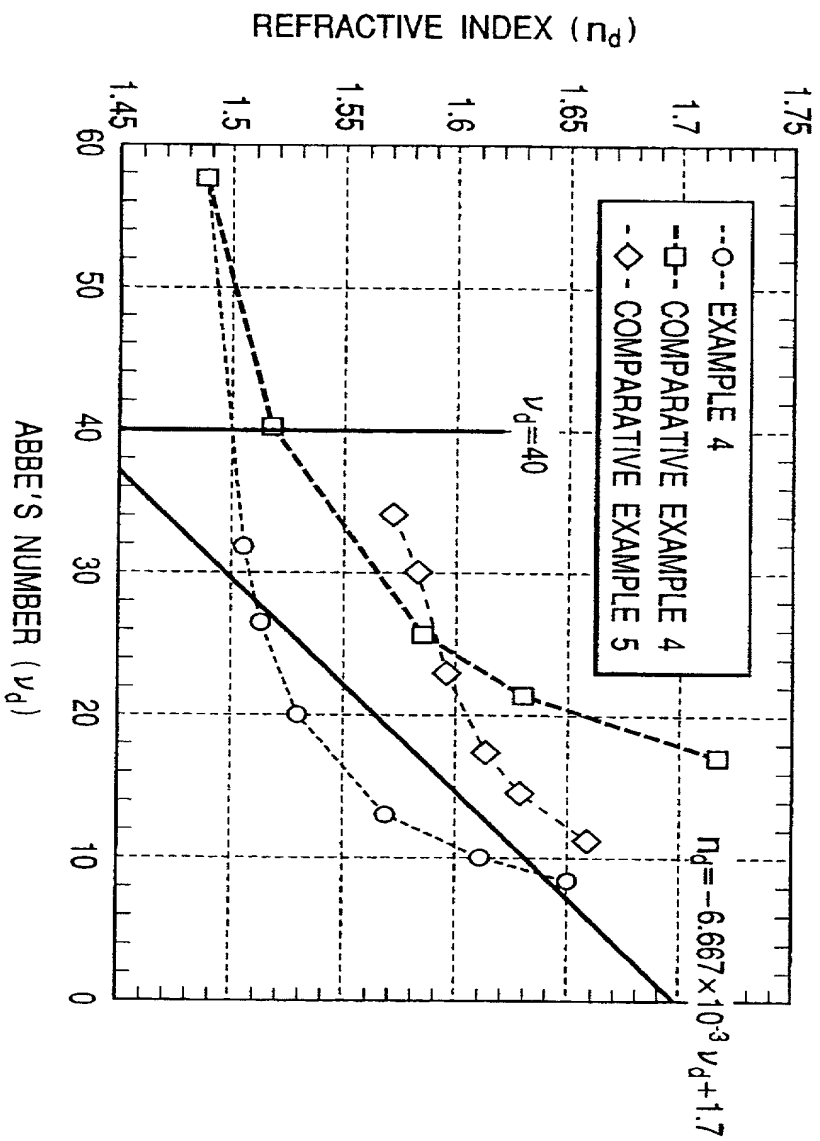


FIG. 13

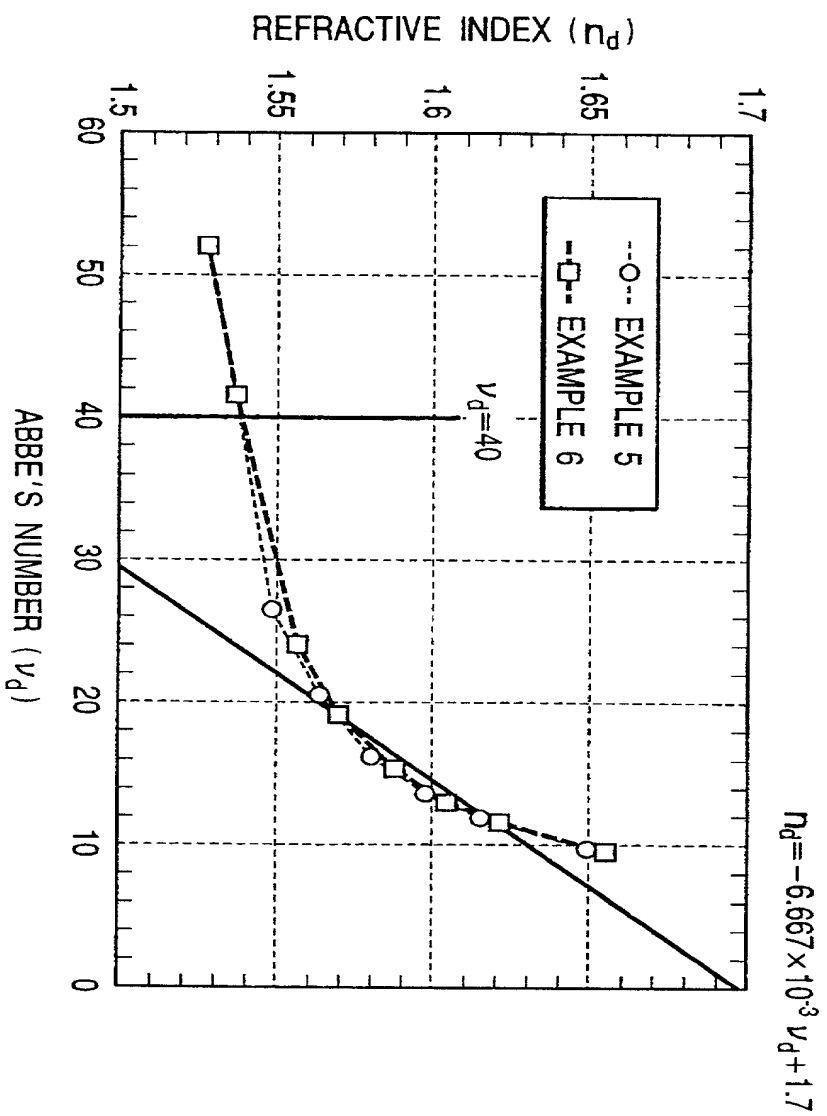
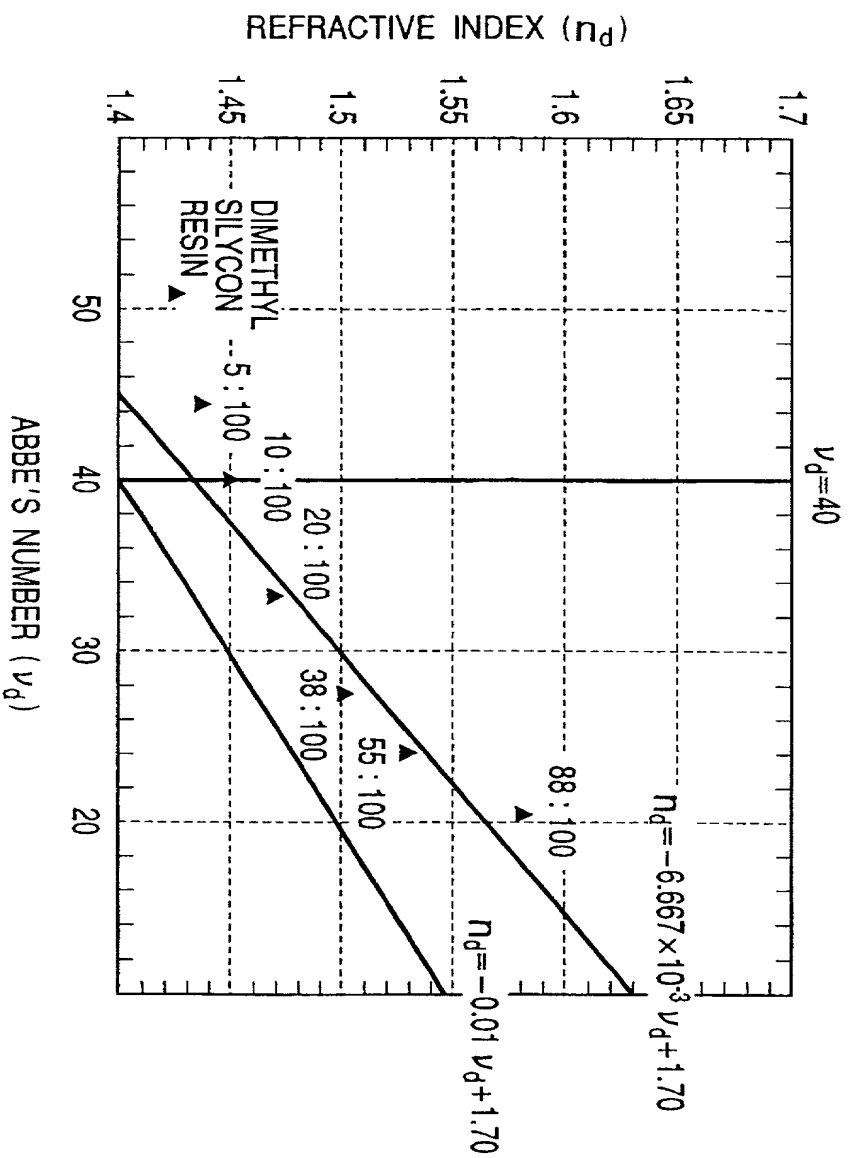


FIG. 14



**COMBINED DECLARATION AND POWER OF ATTORNEY  
FOR PATENT APPLICATION  
(Page 1)**

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled OPTICAL MATERIAL  
AND OPTICAL SYSTEM USING IT the specification of which ☒ is attached hereto ☐ was filed on \_\_\_\_\_  
as United States Application No. or PCT International Application No. \_\_\_\_\_  
and was amended on \_\_\_\_\_ (if applicable).

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR §1.56.

I hereby claim foreign priority benefits under 35 U.S.C. §119(a)-(d) or §365(b), of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT international application which designates at least one country other than the United States, listed below and have also identified below any foreign application for patent or inventor's certificate, or PCT international application having a filing date before that of the application on which priority is claimed:

Country	Application No.	Filed (Day/Mo./Yr.)	(Yes/No) Priority Claimed
JAPAN	11-187678	01 JULY 1999	Yes
JAPAN	12-042600	21 FEBRUARY 2000	Yes

I hereby claim the benefit under 35 U.S.C. § 120 of any United States application(s), or § 365(c) of any PCT international application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT international application in the manner provided by the first paragraph of 35 U.S.C. § 112, I acknowledge the duty to disclose information which is material to patentability as defined in 37 C.F.R. § 1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application.

Application No.	Filed (Day/Mo./Yr.)	Status (Patented, Pending, Abandoned)
-----------------	---------------------	---------------------------------------

I hereby appoint the practitioners associated with the firm and Customer Number provided below to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith, and direct that all correspondence be addressed to the address associated with that Customer Number:

**FITZPATRICK, CELLA, HARPER & SCINTO**  
**Customer Number: 05514**

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Full Name of Sole or First Inventor Hideo UKUDA

Inventor's signature \_\_\_\_\_

Date \_\_\_\_\_ Citizen/Subject of Japan

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